The role of carbon from recycled carbonated metapelites in the origin of ultrapotassic igneous rocks in the Central Mediterranean

Pubrished on Lithos (2015), vol. 232, pag. 174-196 - 10.1016/j.lithos.2015.07.002
Paginated version available at http://www.sciencedirect.com/science/article/pii/S002449371500242X

The role of carbon from recycled sediments in the shift from lamproite to leucitite in the Central Mediterranean region

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Highligths

- lamproites, leucitites and kamafugites from the Central Mediterranean region
- trace elements geochemistry and Sr-Nd-Pb isotope of subduction-related ultrapotassic rocks
- shift from leucite-free to leucite-bearing magmas
- carbonatitic vs. silica-rich metasomatim in the genesis of subduction-related potassic rocks
- slab tears and mantle inflow in the gensis of post-subduction magmas

Abstract

The Central Mediterranean region is one of the most important areas on Earth for studying subduction-related potassic and ultrapotassic magmatism, derived from partial melting of the metasomatised lithospheric mantle wedge. In this region, leucite-free (i.e., lamproite) and leucite-bearing (i.e., kamafugite, leucitite, and plagioleucitite) ultrapotassic rocks closely occur, in a time-related progression, linked to the evolution of both the mantle source and the regional tectonic regime. Time- and space-related magmatism migration followed the roll-back of the subducting slab and the anticlockwise drift of the Italian Peninsula. Leucite-free silica-rich lamproites are restricted to the early stage of magmatism and are associated with ultrapotassic shoshonites and high-K calc-alkaline volcanic rocks. Leucite-bearing (i.e., *Roman Province*) rocks are erupted consistently later than lamproite-like and associated shoshonitic rocks, with post-leucititic volcanism occurring in the late stage of volcanic activity with eruption of alkali-basaltic to latitic and trachytic rocks, often after major caldera-forming events. Present-day ultrapotassic volcanism is restricted to the Neapolitan area. Central Mediterranean potassic and ultrapotassic rocks are extremely enriched in incompatible trace elements with variable fractionation of Ta, Nb, and Ti in comparison to Th and large ion lithophile elements (LILE). They are also variably enriched in radiogenic Sr and Pb and unradiogenic Nd. The main geochemical and isotopic signatures are consistent with sediment recycling within the mantle wedge via subduction. A twofold metasomatism, induced by the recycle of pelitic sediments and dehydration of lawsonite-bearing schists generates the early metasomatic events that enriched the mantle wedge from which leucite-free ultrapotassic rocks (i.e., lamproite) were generated. Recycling of carbonate-rich pelites played an important role in the shift to silica-undersaturated ultrapotassic rocks (kalsilite- and leucite-bearing) of the classic 'Roman province'.

Key words: mantle metasomatism; recycled pelitic to carbonate sediments; ultrapotassic rocks; lamproite, leucitite, kamafugite; Corsica, Tuscany and Roman magmatic Provinces and Central Italy

Introduction

Despite their scarcity, ultrapotassic volcanic rocks are the most studied igneous rocks on Earth because of their peculiar mineralogy, geochemistry, geologic settings and economic value (e.g., Mitchell and Bergman, 1991; Mitchell, 2006). Ultrapotassic igneous rocks originate from partial melting of metasomatised upper mantle sources, in which K-bearing phases have been stabilised by the reaction of surrounding peridotite with K-rich metasomatic melts (e.g., Edgar, 1987, and references therein; Foley, 1992a, 1994). These rocks were initially considered related to intraplate continental settings (e.g., Cundari, 1980), although potassium enrichment is considered an important characteristic of arc-related magmas (e.g., Ninkovich and Hays, 1972; Cawthorn, 1977; De Astis et al., 2000; Francalanci et al., 2004, 2007; Tommasini et al., 2007). Experimental geochemical studies have shown that silica- and potassium-rich magmas can originate at depth, from the recycling of sediments at depths within the upper mantle (e.g., Elliott et al., 1997, 2003; Plank and Langmuir, 1993, 1998; Skora and Blundy, 2010; Avanzinelli et al., 2012a). Geochemical and isotopic studies have also shown that several potassic and ultrapotassic igneous rocks are related to the continental collision, which occurred after closure of the Tethys Ocean, with recycling of upper crust within the sub-continental lithospheric mantle wedge playing a major role (e.g., Cox et al., 1976; Rogers et al., 1985, 1987; Peccerillo, 1985, 1999; Peccerillo et al., 1987; Conticelli and Peccerillo, 1992; Conticelli et al., 2002, 2007; Duggen et al., 2005, 2008; Guo et al., 2006, 2013; Prelević and Foley, 2007).

Despite this large body of work made, limited attention has been paid to the origin of the different types of ultrapotassic magmas ranging from leucite-free, leucite-bearing and kalsilite-bearing ultrapotassic rocks (e.g., Yoder, 1986; Edgar, 1987; Foley et al., 1987). In Italy all these ultrapotassic rock types cluster together in space but not in time, providing a unique opportunity to investigate the mechanisms that control the formation of ultrapotassic magmas with different degrees of silica saturation (Conticelli et al., 2002, 2007).

In this paper we review the trace element geochemistry, the isotopic characteristics, and the petrological constraints of the Central Mediterranean ultrapotassic and associated rocks, in order to better understand: i) the evolution of the mantle source ii) the possible geochemical and isotopic relationships with changes in the geodynamic system and iii) the shift from silica-saturated (leucite-free) to silica-undersaturated (leucite- and kalsilite-bearing) magmatic rocks.

Chronological and geologic outline

In a pioneering study Washington (1906) grouped the potassic and ultrapotassic volcanic rock associations occurring in Italy into three different magmatic '*regions*' on the basis of mineralogical and petrographic characteristics: the Tuscany, Roman, and Apulian regions. The author did not use any temporal constraints, nor did he include or describe the scattered potassic rock cropping out in Corsica, in Tuscany Archipelago, and in intra-pennine areas (Fig. 1). Avanzinelli et al. (2009) recently proposed a classification of Central Mediterranean potassic to ultrapotassic igneous associations, in four magmatic events (i.e., *provinces*; Tab. 1) based on their timing, petrography, and spatial constraints, according to criteria suggested by Turner and Verhoogen (1960) and Conticelli et al. (2004).

The westernmost and oldest outcrops of ultrapotassic leucite-free (i.e., lamproitic) to high-K calc-alkaline volcanic rocks are found in the so-called *Corsica magmatic province* (Fig. 1), along with igneous rocks of Cornacya, Sisco, and Capraia Island (e.g., Peccerillo et al., 1987, 1988; Mascle et al., 2001; Chelazzi et al., 2006; Gagnevin et al., 2007). These rocks were emplaced during the Miocene and yields radiometric ages of 14.3-7.1 Ma (Fig. 2; Civetta et al., 1978; Mascle et al., 2001; Gasparon et al., 2009).

Between the Lower Pliocene (Zanclean) and Lower Pleistocene (i.e., 4.5-0.88 Ma; see Peccerillo, 2005, and references therein) volcanism shifted eastward to the western shoreline of the Italian Peninsula (Fig. 1). This maintained a close spatial association of ultrapotassic leucite-free (lamproitic) with shoshonitic and calc-alkaline suites (e.g., Peccerillo et al., 1987, 1988, 1999, 2005; Clausen and Holm, 1990; Conticelli and Peccerillo, 1992; Conticelli et al., 1992, 2001, 2009a, 2011, 2013). This Plio-Pleistocene magmatic event produced the *Tuscany magmatic province* (e.g., Washington, 1906; Conticelli et al., 2004, 2010; Avanzinelli et al., 2009).

Coeval intrusive to volcanic silicic rocks, which were ultimately derived by anatexis of continental crust, were emplaced in southern Tuscany and within the Tuscany archipelago (Fig. 1) between 6 and 2 Ma. These rocks were kept isolated from the subcrustal igneous rocks of the classic *Tuscany magmatic province* due to their different sources. However, in some cases, hybridisation between mantle and crustal derived magmas has been recorded (e.g., Peccerillo et al., 1987; Poli, 1992; Conticelli et al., 2013).

Following a brief hiatus, magmatism resumed in the Central Mediterranean region during the middle Pleistocene (0.7–0.2 Ma). Ultrapotassic kalsilite- to leucite-bearing igneous rocks were produced at several silica-undersaturated volcanoes of the *Roman magmatic province* (e.g., Washington, 1906; Conticelli et al., 2004, 2010; Peccerillo, 2005; Avanzinelli et al., 2009). This province extends from Monte Amiata to the Neapolitan area (Fig. 1). It runs along as a narrow volcanic belt between the western Italian Peninsula shoreline and the Apennine chain (Fig. 1), showing a broad southeastward migration with time (Fig. 2). At about 400 ka, magmatic activity jumped to a more central region of the Apennine chain, in the Umbria region, marked by the eruption of strongly silica-undersaturated kalsilite-bearing rocks (i.e., kamafugites; Fig. 1). However, no further activity has been recorded in this region (e.g., Gallo et al., 1984; Peccerillo et al., 1987, 1988; Conticelli and Peccerillo, 1992; Stoppa and Cundari, 1998; Conticelli et al., 2004, 2010; Peccerillo, 2005).

The most abundant rocks of the *Roman magmatic province* are silica-undersaturated leucitites and plagio-leucitites (Tab. 1). When present kalsilite-bearing ultrapotassic igneous rocks are confined to early volcanic stages (e.g., Vulsinian volcanoes, the Alban Hills volcano, and the Middle Latin Valley volcanoes)(e.g., Boari and Conticelli, 2007; Boari et al., 2009a, 2009b; Conticelli et al., 2010), and to the intra-Appennine region (i.e., the monogenetic San Venanzo and Cuppaello volcanoes). Hybrid magmas produced by the mixing differentiated leucite-free magmas of the *Tuscany magmatic province* and Roman-type silica-undersaturated magmas, were also erupted during the early stages of activity at some volcanoes (e.g., Amiata, Vulsini, and Vico) where the magmatic provinces overlap (e.g., Cioni et al., 1987; Ferrari et al., 1996; Perini et al., 2000, 2003, 2004; Conticelli et al. 2015; Marroni et al. 2015; Laurenzi et al. 2015). The eruption of *Roman* silica-undersaturated magmas ceased almost coevally during the middle Pleistocene with the eruption of several ignimbrites. These eruptions formed caldera depressions in the *Roman* composite volcanoes during the interval between ca. 300 and 100 ka (e.g., Conticelli et al., 2010).

In some *Roman* volcanoes, during post-caldera activity, pulses of fresh mildly potassic to sub-alkaline leucite-free mafic magmas either replaced or accompanied strongly differentiated leucite-bearing magmas (e.g., Conticelli et al., 1991, 2009b; Perini et al., 2004; Rouchon et al., 2008; Nikogosian and van Bergen, 2010). In the Middle Latin Valley

(Fig. 1), which is devoid of a large composite volcano volcanic activity younger than 300 ka was characterised by the eruption of mildly potassic to sub-alkaline mafic magmas (e.g., Boari and Conticelli, 2007; Frezzotti et al., 2007; Boari et al., 2009a). In the Neapolitan area, the volcanic activity of the *Roman magmatic province* continued into the Holocene. In this region (i.e., Neapolitan district; Washington, 1906) a cluster of four composite volcanoes (i.e., Ischia, Procida, Phlegrean Fields and Somma-Vesuvius volcanoes; Fig. 1) produced large pyroclastic and lava eruptions in the last 300 ka, which have occurred through to the present time with intense historical volcanic activity (e.g., Poli et al., 1987; Orsi et al., 1995; Civetta et al., 1997; D'Antonio et al., 1999a, 1999b, Pappalardo et al. 1999, 2002; Brocchini, et al., 2001). In the Neapolitan area, apart from few leucite-free mafic rocks, trachytic to latitic rocks have also been erupted (Fig. 2) along with the few mafic leucite-free mafic rocks (e.g., Peccerillo, 2005, and references therein). The production of leucite-bearing magmas resumed at Vesuvius during the Holocene and has continued to the present day, with the last eruption occurring in 1944 A.D. (e.g., Peccerillo, 2005 and references therein; Cioni et al., 2008).

The *Lucanian magmatic province* is the southeastern most volcanic region on the Italian Peninsula (Fig. 1), the so-called 'Apulian region' of Washington (1906). *Lucanian* volcanic activity gave rise to the Vulture and the Monticchio nested volcanoes, along with a few small and scattered explosive monogenetic centres along the Ofanto Valley (authors' unpublished data). Mildly potassic to sodic volcanic rocks were erupted between 740 and 130 ka during the upper Pleistocene (Fig. 2) (e.g., Villa and Buettner, 2009), with a final carbonatitic lava flow occurring at Toppo del Lupo (e.g., De Fino et al., 1986; Melluso et al., 1996; Bindi et al., 1999; Rosatelli et al., 2000; Beccaluva et al., 2002; De Astis et al., 2006; D'Orazio et al., 2007, 2008; Stoppa et al., 2008). The Vulture alkaline rocks and carbonatites show intermediate characteristics between subduction-related and intraplate volcanic rocks. This hybrid nature is due to the occurrence of a slab tear beneath the Vulture area, which allowed the inflow of an intraplate component into the Italian sub-continental mantle wedge (e.g., Faccenna et al., 2007; Avanzinelli et al., 2008; Rosembaum et al., 2008). Given the mostly sodic rather than potassic nature of *Lucanian* rocks we consider them outside the scope of this study and thus they are not considered further.

Materials and Methods

The discussion reported in the following paragraphs is based on collection of data reported in the supplementary materials. Samples selection has been performed to cover the entire compositional spectrum of the Central Mediterranean ultrapotassic magmatism. Samples with clear signs of secondary alteration, where possible, were excluded. In addition, to ensure consistency within the data set, analytical methods, precision, and accuracy were evaluated to minimise bias toward samples from certain laboratories. Major element, trace element, and isotope data for upper crustal rocks and sediment from the Central Mediterranean region are also reported along with data from the literature in the supplementary materials, which are available for downloading as excel files from the journal's website.

A subset of samples was extracted from the whole data set in order to discuss the primary characteristics of the magmas. As such, when discussing the geochemical and isotopic signatures of mantle sources only the samples thought to represent the composition in equilibrium with mantle region, or near to it, were considered. With this in mind, samples in the subset were selected according to the following criteria: i) MgO and [MgO]/([MgO]+[0-85*Feo_{tot}]) (i.e., Mg#) higher than 4 wt.% and 0.6, respectively; ii) low porphyritic index (P.I.) to ensure that chemical constraints on Mg/Fe and compatible trace elements were not governed by cumulative mafic phases; and iii) euhedral olivine formed in the melt with a composition in equilibrium with that of the bulk rock. In table 1 main mineralogic, petrographic, and chemical features of the different groups recognised are also reported.

Ultrapotassic suites

In the total alkalis *vs*. silica diagram (Fig. 3A), the Central Mediterranean potassic and ultrapotassic igneous rocks fall in nearly all the alkaline fields above the alkali divide, (Kuno, 1968) ranging from high-K calc-alkaline to shoshonitic, and ultrapotassic (Fig. 3B), although some also plot in the high-K calc-alkaline field. Magmatic differentiation is responsible for the chemical evolution seen within each magmatic series (Fig. 3). Liquid lines of descend in the Mg# *vs.* silica diagram show two broad parallel arrays (Fig. $3C_1$). Leucite-free ultrapotassic to potassic rocks of the *Corsica* and *Tuscany magmatic provinces* (Miocene to Lower Pleistocene) have higher SiO_2 contents than the younger (upper Pleistocene to Holocene) leucite-bearing ultrapotassic rocks and later leucite-free rocks (herein termed 'post-leucitites') of the *Roman magmatic province* (Fig. $3C_1$).

The nomenclature and classification of potassic and ultrapotassic rocks have long been debated because of the complex mineralogy and extreme chemical variability of the magmas (e.g., Barton, 1979; Mitchell and Bergman, 1991; Wolley et al., 1996; Frost and Frost, 2008). The mineralogical and chemical variabilities may result from i) incomplete reactions between mineral phases with narrow stability ranges (i.e., heteromorphism; Yoder, 1986), ii) different X_{CO2} conditions and total pressure during partial melting (e.g., Wendlandt and Eggler, 1980a, 1980b; Conticelli, 1989; Foley, 1992a, 1994); iii) different mineralogy of the mantle source region (e.g., Edgar, 1987). A unified nomenclature used to differentiate the different groups of primary ultrapotassic magmas does not yet exist; however, Le Maitre (2002) suggested using the scheme of Foley et al. (1987) (Tab. 1, Fig. 4).

In this study the rocks of the Central Mediterranean region have been divided according to their geography, mineralogy and geochemical character, into two main groups: the *Corsica* and *Tuscany magmatic provinces* and the *Roman magmatic province*. Each group is further subdivided according to key geochemical features described by Avanzinelli et al. (2009). It is worth highlighting that lamproites display a high silica content (up to 60%) despite maintaining a 'primitive' character, as shown by their high MgO contents and Mg# (Fig. 3), and by the almost ubiquitous occurrence of highly forsteritic (Fo_{85}-Fo_{95}) olivine, which is in chemical equilibrium with the bulk rocks.

Corsica and Tuscany magmatic provinces

Lamproites from the *Corsica* and *Tuscany magmatic provinces* (dark green circles in Fig. 3) are silica-saturated to silica-oversaturated (i.e., *hypersthene* normative; Tab. 1, Fig. 5A, and supplementary data), with high-MgO levels, ranging in composition from lamproite (orendite) to lamprophyre (minette)(Rock, 1991); differentiated felsic compositions are rarely found (e.g., Gallo et al., 1984; Peccerillo et al., 1988; Conticelli et al., 1992; Conticelli, 1998). Lamproites are invariably plagioclase-free and leucite-free ultrapotassic rocks, characterised by highly forsteritic olivine, chromian spinel, Al-poor clinopyroxene, K-richterite, sanidine, picro-ilmenite, and apatite (Tab. 1). Leucite may occur in silica-undersaturated lamproites, but they have never been observed within the Central Mediterranean lithologies (e.g., Wagner and Velde, 1986; Mitchell and Bergman, 1991; Conticelli et al., 2009a).

Ultrapotassic shoshonitic rocks from the *Corsica* and *Tuscany magmatic provinces* (pale green circles in Fig. 3 and following) are *hypersthene*- to *quartz*-normative rocks (see supplementary data) with a positive feldspathoid silica-saturation index (FSSI, Frost and Frost, 2008). They can be distinguished from lamproite by their higher CaO, Na_2O,

Al_2O_3.,and lower K_2O contents (Tab. 1), as well as their lower peralkaline index (Fig. 5A). These chemical characteristics mean that the *Corsica* and *Tuscany* shoshonites, although still ultrapotassic (Fig. 5B), do not consistently fall within one field on the discrimination diagrams for ultrapotassic rocks (Fig. 4), but are well within the field of shoshonites in the K_2O *vs.* Silica plot (Fig. 3B). These rocks are also characterised by variable MgO and silica contents depending upon their degree of differentiation (e.g., Conticelli et al., 2009a). They are leucite-free with abundant plagioclase usually restricted to the groundmass, although it is rarely found as phenocrysts subordinate to olivine and clinopyroxene. Chromian spinel is found in the core of olivine phenocrysts. Sanidine, magnetite, and apatite are restricted to the groundmass. Orthopyroxene is found in the shoshonite samples with intermediate silica contents (Conticelli et al., 2013) and among high-K calc-alkaline rocks (Chelazzi et al., 2006; authors' unpublished data).

Roman magmatic province

Kamafugites from *Latium* and *Umbria* (Fig. 1)(red diamonds in Fig. 3) are rare kalsilite-bearing melilitolites, named after those first described by Holmes and Harwood (1932) in Central Africa. They are strongly silica-undersaturated ultrapotassic rocks (i.e., *larnite* normative, see supplementary data; Tab. 1, Fig. 5A), with high MgO and CaO contents, although intermediate varieties do also occur. Central Mediterranean kamafugites are feldspar-free and consist of felsic phases including kalsilite, nepheline and rare leucite along with mafic phases, such as olivine, Al-poor clinopyroxene, phlogopite, and melilite. Perovskite, ilmenite, and apatite occur as accessory phases. The Central Mediterranean is the only region in which these rocks are intimately associated with other types of ultrapotassic rocks (Conticelli and Peccerillo, 1992; Cellai et al., 1994). Carbonatites in Central Italy have also been repeatedly claimed to occur in association with kamafugitic melilitolites (e.g., Stoppa and Cundari, 1995, 1998; Stoppa and Wolley, 1997), although no rocks with modal calcite have yet been unequivocally found in the *Roman magmatic province*. Debate surrounding this issue is ongoing (e.g., Peccerillo, 1998; Melluso et al., 2003, 2005a, 2005b; Martin et al., 2012).

Leucitites and plagio-leucitites (orange diamonds in Fig. 3) are restricted to the volcanoes of the *Roman magmatic province* (e.g., Washington, 1906; Conticelli et al., 2004; Avanzinelli et al., 2009). These rocks are exclusive to the cone building phases of the main volcanic complexes of the province (e.g., Vulsini, Vico, Sabatini, and Roccamonfina volcanoes), as well as the oldest products of the monogenic volcanoes of the Middle Latin Valley. However, they are also found in the final activity of Vesuvius, after formation of the Monte Somma caldera (Fig. 1)

The leucitites are mostly plagioclase-free whilst plagio-leucitites have modal plagioclase alongside leucite as the major felsic phases. Leucitites and plagio-leucitites have highly variable degrees of silica undersaturation (Fig. 5A), but with SiO_2 rarely exceeding 65 wt.% as seen in phonolites. They are characterised by extremely variable MgO contents, relatively low FeO_{tot}, and relatively high CaO, Al_2O_3, and Na_2O contents. Leucitites close to the peralkaline divide are plagioclase-free (Fig. 5A). Olivine and Al-rich clinopyroxene are the most abundant mafic phases; phlogopite is found in the groundmass of mafic leucitites and tephrites and biotite in the phonolite. Magnetite, nepheline, sanidine, sphene, and apatite are restricted to the groundmass. In contrast to lamproites and kamafugites, clinopyroxene in leucitites and plagio-leucitites is Al-rich, with Al completely filling the deficiency of silicon in the tetrahedral site (Cellai et al., 1994), while high-forsterite olivine is enriched in CaO compared wile olivine from lamproites and shoshonites (Boari and Conticelli, 2007; Ammannati et al., 2015).

Young post-leucititic basalts to trachytes (dark yellow diamonds in Fig. 3) occur in the post-caldera phases of some *Roman* volcanoes and in the youngest products (<300 ka) of the monogenic volcanic field of the Middle Latin

Valley (Fig. 1). These rocks are also typical of most of the volcanoes of the Neapolitan area (e.g., Ischia, Procida, Phlegrean Fields, and Somma volcanoes, Fig. 1), with the exception of Vesuvius (e.g., Beccaluva et al., 1991; Civetta et al., 1997; Conticelli et al., 2004, 2009b; D'Antonio et al., 1999a,b; Pappalardo et al., 1999, 2002; Peccerillo, 2001; Cioni et al., 2008). Young post-leucititic basalts to trachytes are silica-saturated to slightly silica-undersaturated (Fig. 5B), mildly enriched in potassium, and subordinate in volume to older leucite-bearing rocks when occurring in the Latium volcanoes (i.e., Vulsini, Vico, Middle Latin Valley, and Roccamonfina volcanoes; Fig. 1). The petrography and mineralogy of these post-leucititic rocks closely resemble those of the shoshonite and calc-alkaline suites of the older *Corsica* and *Tuscany provinces* (Tab. 1), although orthopyroxene is missing in these young mildly potassic to sub-alkaline rocks (Fig. 3)(e.g., Conticelli et al., 2004, 2009b; Fedele et al., 2006; Boari and Conticelli, 2007; Melluso et al., 2012).

Figure 4 shows diagrams for discriminating among different types of ultrapotassic rocks; post-leucititic rocks are not plotted because they are not ultrapotassic (Fig. 5B). Leucite-free ultrapotassic rocks of the *Corsica* (i.e., Sisco) and the *Tuscany* (i.e., Orciatico, Montecatini val di Cecina, and Torre Alfina) *magmatic provinces* plot uniformly within the lamproite field, with few samples falling on the boundaries (Fig. 4). Shoshonites of the *Tuscany magmatic province* (e.g., Radicofani, Torre Alfina, and Cimini volcanoes) do not fall unequivocally in a single ultrapotassic field (Fig. 4). Kalsilite- and leucite-bearing ultrapotassic rocks of the *Roman magmatic province* fall uniformly within the kamafugite and leucitite & plagio-leucitite fields, respectively (Fig. 4).

A more useful distinction between the Italian potassic and ultrapotassic rocks can be made on the basis of silica saturation. The oldest mafic high-MgO (Mg# > 0.6) rocks belonging to the *Corsica* and *Tuscany magmatic provinces* have feldspathoid silica-saturation index (FSSI) > 0, while those belonging to the classic *Roman magmatic province* have FSSI < 0 (Fig. 5A). In the *Roman magmatic province,* the FSSI broadly correlates with potassium enrichment, as the rock types_most enriched in K_2O (i.e., kamafugites, leucitites, and plagioleucitites) show the most silica-undersaturated compositions, while the post-leucitites plot closer to the silica saturation line (Fig. 5A).

Almost all of the mafic high-MgO igneous rocks (Mg# >0.6) that erupted during the early magmatic activity in the Central Mediterranean, are ultrapotassic (*Corsica* and *Tuscany magmatic provinces*). In contrast among the *Roman* rocks only kamafugites, leucitites, and plagio-leucitites are ultrapotassic. Mafic high-MgO post-leucitites (i.e. basalts) plot below the potassic/ultrapotassic divide in the Total Alkali Silica (TAS) diagram (Fig. 5B).

Role of the Continental Crust and the Plume hypothesis

The involvement of the continental crust in the genesis of Mediterranean ultrapotassic rocks is clear. Many geochemical and petrographic tracers highlight the role of continental crust, either in the genesis of the primary melts or during the shallow level differentiation of the magmas. Many studies have revealed the role of crustal assimilation combined to fractional crystallisation (i.e., AFC) in driving mafic parental magmas to low MgO and high silica compositions in their individual potassic suites (e.g., Peccerillo et al., 1984; Conticelli et al., 1987, 1991, 1992, 1997, 2001, 2009b, 2013; Civetta et al., 1997; Pappalardo et al., 1999, 2002; Perini et al., 2004; Boari et al., 2009a, 2009b). However the key geochemical and isotopic characteristics of the studied igneous rocks, such as the extreme enrichment in potassium and incompatible trace elements, and the exotic radiogenic isotope compositions of the least differentiated magmas from each volcano, cannot be ascribed to shallow-level differentiation processes such as the assimilation of continental crust (Peccerillo 1989, 2005; Conticelli et al., 2002, 2007).

Previous studies have thoroughly discussed this issue, showing that in high-MgO (mafic) ultrapotassic rocks, with olivine in compositional equilibrium with the bulk rock, crustal contamination en route to the surface is negligible (e.g., Conticelli, 1998; Conticelli et al., 2002, 2007, 2011, 2013; Murphy et al., 2002; Prelević et al., 2008, 2010; Perini et al., 2004; Boari et al., 2009a, 2009b). In the following points, we outline some key arguments against a major role for crustal contamination in controlling the chemical and isotopic composition of the magmas that formed beneath the Italian Peninsula.

i) All the selected samples are mafic (Mg# > 0.6, Fig. 3C$_2$) and have aphiric to sub-porphiric textures with high-forsterite (Fo_{95}-Fo_{85}) liquidus olivine in equilibrium with the bulk rock composition. Crustal contamination is a heat-consuming process that would have resulted in the rapid crystallisation and fractionation of mafic minerals, taking highly forsteritic olivine away from the liquidus of the magma;

ii) Despite a large variation in the whole isotope dataset, there is no correlation between common geochemical discriminants and Nd or Sr isotopes, which would be expected if AFC processes had occurred. The isotope ratios remain distinguishable among different provinces even with similar levels of differentiation and, most importantly, little variation in the Nd and Sr isotope ratios is observed within each province at increasing levels of magma differentiation (Conticelli and Peccerillo, 1992);

iii) Given the extreme enrichment of incompatible trace elements in high-MgO and olivine-bearing ultrapotassic magmas (twice that of any crustal rock), the occurrence of crustal contamination would dilute, rather than enrich, the composition of the resulting magmas (e.g., Conticelli, 1998; Murphy et al. 2002);

iv) Recent studies on solid-melt inclusions in highly forsteritic (Fo_{85}-Fo_{95}) olivine (which is in equilibrium with high-MgO mafic Italian potassic and ultrapotassic rocks) show that both the trace elements (Schiano et al., 2004; Nigokosian and van Bergen 2010) and the isotopic composition (Koornneef et al., 2015), of the melt inclusions are indistinguishable from those of the bulk rock. As such, melts isolated within highly forsteritic olivine should represent the composition of the magma before any potential crustal assimilation process occurred.

With these points in mind, we conclude that most of the residual geochemical and isotopic crustal signatures in the bulk rock compositions of the selected samples must be considered as primary characteristics acquired from the mantle-sources of these magmas.

Incompatible trace elements in mafic ultrapotassic rocks

The most primitive ultrapotassic igneous rocks of the Central Mediterranean region show high concentrations and fractionated contents of incompatible trace elements (Fig. 6). Primitive-mantle-normalized spider diagrams of mafic ultrapotassic and potassic igneous rocks show extreme but variable enrichments in large ion lithophile (LILE) elements (i.e., Th, Hf, and Zr) with respect to other high field strength (HFSE) elements (i.e., Ta, Nb, P, and Ti; Fig. 6). These trace element characteristics are similar to those of sediment and continental upper-crustal rocks (Fig. 7A–C). A strong Pb peak is thought to be a distinctive feature of crustal material that has been assimilated into the upper mantle. Conversely, spider diagrams of the mafic Central Mediterranean ultrapotassic rocks (Fig. 6) show large differences when compared with those of intraplate ultrapotassic rocks and carbonatites (Fig. 7D). They do not show troughs in Ba, Nb, Ta, Hf, and Ti, or peaks in U, Th, and Pb. Carbonatites associated with ultrapotassic rocks show an additional trough at K (Fig. 7D).

The pioneering geochemical studies of Cox et al. (1976) and Rogers et al. (1985, 1987) argued that potassic magmas are derived from an anomalously K-enriched mantle. More recently it has also been shown that fractionation of HFSE, with respect to LILE, is a typical characteristic of subduction-related magmatic suites that are derived from the partial melting of an anomalously enriched mantle wedge (e.g., Rogers et al., 1987; Plank and Langmuir, 1993, 1998; Elliott, et al., 1997; Elliott, 2003; Avanzinelli et al., 2012a).

Isotopic composition of mafic ultrapotassic rocks

The isotopic ratios of conventional and unconventional isotopes (e.g., Sr, Nd, Pb, Hf, He, Os, U, and Th) in mafic high-MgO ultrapotassic rocks of the Central Mediterranean region support the hypothesis that the continental crust played a major role in their genesis (e.g., Conticelli et al., 2002, 2007; Gasperini et al., 2002; Martelli et al., 2004; Avanzinelli et al., 2008).

Even after the accurate selection of the most mafic samples (Mg#> 0.6 with MgO > 4 wt.%), Italian ultrapotassic mafic rocks still show negatively correlated Nd and Sr isotopic compositions (Fig. 8A), which could potentially be ascribed to shallow level crustal contamination. Koornneef et al. (2015) reported that solidified melt inclusions in olivine from Italian high-Mg ultrapotassic rocks are consistent with the Nd and Sr isotopic compositions observed in the bulk rock, indicating the melt inclusions are in isotopic equilibrium with their hosts (Fig. 8A). With crustal contamination being a heat-consuming process, massive fractionation of mafic phase should have occurred (especially for the extreme amount of crustal contaminant needed up to 75 vol.%; Ferrara et al., 1986) in order to drive the Nd and Sr isotopic composition to the most radiogenic and non-radiogenic Sr and Nd, respectively, thereby matching the values observed in lamproites (Fig. 8A). Such a large amount of crustal contamination would have i) dramatically decreased the compatible trace element content, which is not observed in our ultrapotassic and associated mafic samples (Conticelli and Peccerillo 1992; Nelson 1992; Murphy et al., 2002; Prelević et al., 2008; Tommasini et al., 2011); and ii) produced a direct correlation between magma differentiation and crustal the isotopic signature, which is not observed in the whole Western Mediterranean lamproite samples (Conticelli et al., 2009a). In addition, for simple mass balance reasons, a relatively small amount of crust-derived melt, recycled into a previously depleted mantle wedge, would be sufficient to explain the isotope compositions of the Italian magmas (<5% for the *Roman*-type magmas; Avanzinelli et al., 2008).

Mixing between lamproite-like magmas and felsic crustal-derived magmas is also not a viable process that can explain the isotope composition of the rocks studied here. Given that crustal-derived melts are characterised by higher Nd isotopes than lamproites, such a mixing process would have driven Nd and Sr isotopes toward a positive correlation (Conticelli et al., 2013).

In summary, the Central Mediterranean ultrapotassic and associated mafic rocks terms with Mg# > 0.6 and MgO > 4 wt.%, and their olivine-hosted solid-melt inclusions, are aligned along a mixing array between upper mantle and continental crust end-member in the $^{143}Nd/^{144}Nd_i$ *vs.* $^{87}Sr/^{86}Sr_i$ diagram (Fig. 8A). This array is believed to represent a primary characteristic of their mantle source due to the recycling of felsic melts (produced by the melting of sediments) via subduction within the upper mantle (e.g., Avanzinelli et al., 2008, 2009). A decoupling of the relationship between Sr and Nd isotopes is also observed between leucite-bearing and leucite-free trends, with the *Roman* plagio-leucitites, leucitites, and kamafugites pointing having lower $^{143}Nd/^{144}Nd_i$ values than the rocks of the *Corsica* and *Tuscany magmatic provinces* (Fig. 8A). The northernmost and older potassic rocks plot at the low-radiogenic Nd and high-radiogenic Sr end, while the southernmost and younger post-leucitites plot at the high-radiogenic Nd and low-radiogenic Sr end (Fig. 8A). As a results the mafic ultrapotassic rocks of the *Roman magmatic province* are distinguishable from other provinces using radiogenic isotopes: the leucite-bearing rocks from Latium and Northern Campania (i.e., the Latium districts: Vulsini, Vico, Sabatini, Colli Albani, Middle Latin Valley, and Roccamonfina; Fig. 1) show higher $^{87}Sr/^{86}Sr_i$ and lower $^{143}Nd/^{144}Nd_i$ values than the rocks of the Neapolitan region (i.e., the Neapolitan district made up by the cluster of Somma-Vesuvius, Ischia, Procida, and Phlegrean Field volcanoes; Fig. 1). Post-leucitites from post-caldera phases (i.e., Vulsini, Vico, and Roccamonfina volcanoes) and from the Middle Latin Valley volcanoes show intermediate values between those of the Latium and Neapolitan rocks (Fig. 8A). Notably, the plagio-leucitites of Vesuvius have lower $^{87}Sr/^{86}Sr_i$ and higher $^{143}Nd/^{144}Nd_i$ than similar rocks from the northernmost volcanoes, along with most of the post-leucititic basalts and trachybasalts (Fig. 8A).

Lead isotopes display a composite behaviour with distinctive correlation trends for i) lamproite-like and associated mafic high-Mg rocks (leucite-free) of the *Corsica* and *Tuscany magmatic provinces*, ii) mafic high-MgO kamafugite, leucitite, and plagio-leucitites (kalsilite- to leucite- bearing) of the *Roman province*, and iii) the volcanic rocks of the Neapolitan volcanoes (e.g., Conticelli et al., 2009b, and references therein). In figure 8B ($^{206}Pb/^{204}Pb_i$ *vs.* $^{87}Sr/^{86}Sr_i$), leucite-free and leucite-bearing ultrapotassic rocks define an array between an upper crustal component and an intraplate-like component (similar to the magma compositions of the *Lucanian magmatic province* and those erupted within the Sicily Channel). Conversely, post-caldera Roccamonfina rocks and the youngest post-leucitites of the Neapolitan volcanoes trend toward a MORB-like geochemical reservoir (i.e., depleted mantle; Fig. 8B). For the first trend the most ultrapotassic terms of both leucite-free and leucite-bearing rocks (i.e., lamproite, kamafugite and leucitite) plot toward the upper crustal component for each volcanic suite. For the second trend, we infer the mixing of three isotopic components in the source region of the young Neapolitan rocks (Fig. 8B), comprising the following end-members: 1) MORB-like depleted pre-metasomatised mantle, 2) recycled sediment, and 3) an intraplate geochemical component (Avanzinelli et al., 2009). The intraplate geochemical component is thought to have recently invaded through a slab tears and via lateral mantle flows, the sub-continental mantle wedge beneath southern Italy (e.g., Faccenna et al., 2001; Funiciello et al., 2004; Avanzinelli, 2008, 2009; Rosenbaum et al., 2008; Conticelli et al., 2009b; Bell et al., 2013). The newly arrived geochemical component diluted the original crustal signature of the post-leucititic *Roman* basaltic and trachybasaltic magmas of the Southern Latium areas and possibly those of the Neapolitan area (e.g., De Astis et al., 2006; Conticelli et al., 2009b, 2013).

The co-variation of $^{176}Hr/^{177}Hf_i$, $^{187}Os/^{188}Os_i$, and $^{3}He/^{4}He$ (R/RA) *vs.* $^{87}Sr/^{86}Sr_i$ (Fig. 9) provides further evidence for interaction between a recycled crustal-derived component and an asthenospheric mantle end-member, although the distinction between the roles of MORB-like depleted mantle and the intraplate component is not so evident. A negative correlation between initial Hf and Sr isotopes is observed (Fig. 9A), with leucite-free ultrapotassic rocks (i.e., lamproites and shoshonites) having isotopic values that overlap with those of the upper crusts, as well as having similar geochemical characteristics to those of other Western Mediterranean lamproites (i.e., Western Alps, and Spain; Prelević et al., 2010). Leucite-bearing ultrapotassic rocks (i.e., kamafugites and leucitites) have higher

$^{176}Hr/^{177}Hf_i$ values, intermediate between those of the crustal and the mantle end-members (Fig. 9A). In contrast young Neapolitan rocks plot much closer to the mantle end-member suggesting the arrival of either an intraplate component through slab tear (Gasperini et al., 2002), or a massive fluid-like component (e.g., Avanzinelli et al., 2008).

A similar pattern to those of other radiogenic isotopes is observed for $^{187}Os/^{188}Os_i$ *vs.* $^{87}Sr/^{86}Sr_i$ isotope ratios with data from the leucite-free rocks (lamproite) pointing to a crustal end-member, while the leucite-bearing rocks (kamafugites, leucitites and plagio-leucitites) have lower values (Fig. 9B), which are still consistently higher than those of the upper mantle (e.g., Brandon et al., 1996; Carlson, 2005). Osmium is much more sensitive to shallow-crustal contamination processes than any other isotopic system (e.g., Chesley et al., 2004; Chen et al., 2013) due to the strongly compatible nature of Os, which is much more enriched in the mantle than in the crust. Indeed, the scatter towards high $^{187}Os/^{188}Os$ ratios observed in figure 9B can be ascribed to small amounts of shallow-level crustal contamination. However, the overall isotopic trend shown in figure 9B cannot be explained by crustal assimilation alone, which is consistent with other evidence discussed above (e.g. Sr-Nd isotopes in melt inclusions). Significant amounts of recycled crustal material within the upper mantle are required for the formation of lamproitic and kamafugitic magmas, which become diluted passing to post-leucitites (Conticelli et al., 2007). In addition, the Sr-Os isotope values of Italian and Serbian lamproites plot along a mixing trend between peridotitic and phlogopite-bearing clinopyroxenite xenoliths (Fig. 9B), with the latter thought to represent the mineralogy of the mantle source of leucite-free Western Mediterranean lamproites (e.g., Prelević et al., 2014).

He isotopes of melt inclusions in olivine and clinopyroxene crystals from rocks of the *Tuscany* (lamproites) and *Roman* (leucitites and plagio-leucitites) *magmatic provinces* also indicate a recycled crustal component. This adds a further constraint on the occurrence of a crustal component within the Central Mediterranean sub-continental mantle wedge (Martelli et al., 2004). Overall, the studied samples have significantly lower $^{3}He/^{4}He$ (R/RA) values than MORB, showing a negative correlation between $^{87}Sr/^{86}Sr_i$ and lowest $^{3}He/^{4}He$ (R/RA) found in the most ultrapotassic end-members of each volcanic suite (Fig. 9C).

Importantly, the Sr and Pb isotopic ratios of the plagio-leucitites from Vesuvius overlap with those of the post-leucititic rocks. These data are not plotted here because when MgO < 4wt.%, the Hf (Gasperini et al., 2002) and He (Martelli et al., 2004) isotope data plot closer to the mantle end-member than to the same lithotypes from the northernmost districts (Latium) of the *Roman magmatic province.*

An alternative explanation for the geochemical and isotopic characteriscs observed in the Italian potassic and ultrapotassic rocks involves the presence of a large mantle plume beneath the Central Mediterranean area (Bell et al., 2013). In a recent comprehensive study of U-Th disequilibria in the youngest Central Mediterranean rocks, Avanzinelli et al. (2008) investigated the possibility of ultrapotassic magmas being generated by decompression melting during the upwelling of a deep mantle plume. The presence of significant excess ^{238}U in the volcanic rocks of the Neapolitan district of the *Roman magmatic province* (Fig. 10), strongly argues against the plume hypothesis. In fact, excess ^{238}U is key evidence for the subduction-related genesis of magmas in a volcanic arc (e.g., Elliott et al., 1997; Hawkesworth et al., 1997). The restricted occurrence of excess ^{238}U in the Neapolitan rocks requires the recent addition of a U-rich fluid-like component that is expected to trigger partial melting of the mantle wedge (Avanzinelli et al., 2008). The lack of any significant disequilibrium in the Latium districts of the *Roman magmatic province* (Fig. 10) is also inconsistent with a genesis by adiabatic partial melting of a deep mantle plume. Indeed, partial melting in an upwelling mantle plume is expected to produce significant excess ^{230}Th, as found in a sample from the Monticchio volcano in the *Lucanian magmatic province*. This excess ^{230}Th was interpreted to have resulted from the passive upwelling of the mantle through a slab tear rather to a deep-seated mantle plume (Avanzinelli et al., 2008).

The subduction factory and the 'carbonatite' connection

One important issue in constraining the anomalous potassium enrichment of the upper mantle at destructive plate boundaries is the origin and mechanism of trace element transfer from the slab to the mantle wedge (e.g., Elliott, 2003; Tommasini et al., 2007; Avanzinelli et al. 2012a). Incompatible elements in the source of subduction-related magmas may be derived from either dehydration of altered oceanic crust or recycled sediments and can be transferred within fluid-like and/or melt-like carriers, respectively.

Decoupling of Th and incompatible high field strength elements

Subduction-related ultrapotassic rocks have high Th concentrations, high Th/Nb and Th/REE ratios (Fig. 11), and low Ba/Th (Avanzinelli et al., 2009), which are indicative of recycled sediment within their mantle source as melts rather than aqueous fluids (e.g., Plank and Langmuir, 1993, 1998; Elliott et al., 1997; Avanzinelli et al., 2008, 2009). The enrichment in Th and lithophile incompatible elements with concurrent depletion in HFSE has been modelled experimentally as being due to the presence of residual rutile during the partial melting of sediment within a subduction zone (e.g., Klimm et al., 2008; Skora and Blundy, 2010; Martindale et al., 2013). At temperature of the slab-mantle interface, accessory phases such as rutile along with allanite and/or monazite (and possibly apatite), are stabilised in the residue of recycled sediment during slab melting, strongly sequestering REE and Th (in allanite and/or monazite), or Ti and Nb (in rutile) from the metasomatic liquids. The resulting metasomatic agent is then enriched in Cs, Rb, Ba, K, Pb, Sr, and U relative to Th (i.e., all element not hosted by the residual phases), which reflects the composition of the slab-derived aqueous fluid (Klimm et al., 2008; Avanzinelli et al., 2012). At higher temperatures and pressures metasomatic fluids incorporate further amounts of solutes (i.e., melts) with a concurrent increase in the solubility of allanite and monazite. Full elimination of these two accessory phases from the residue of recycled sediments causes a massive release of Th, U, and REE, into the melts, while rutile remains in the residue (e.g., Hermann, 2002; Kessel et al., 2005; Klimm et al., 2008). The partition coefficients of LREE and Th in allanite and monazite are much higher than those for heavy REE (HREE) and U; consequently, allanite-saturated fluids from recycled pelitic sediment would have lower LREE/HREE and Th/U ratios than their sedimentary protolith. In contrast, if temperatures are high enough to keep the metasomatic liquids undersaturated in allanite, Th, U and REE can be released into the liquids and their concentrations and interelemental ratios would depend upon other mineral phases. The high Th/U and LREE/HREE ratios of most Central Mediterranean ultrapotassic rocks, with the exception of the young post-leucititic Neapolitan rocks (Fig. 11), suggest that the dominant metasomatic agent was a melt rather than a fluid, generated within the stability field of rutile during partial melting of metapelitic sediments, which kept the melt depleted in HFSE but not in Th.

Partial melting of recycled metapelitic sediment with garnet in the residue is also responsible for the high LREE/HREE raio of subduction-related K-rich metasomatic melts (i.e., Kessel et al., 2005; Avanzinelli et al., 2008, 2012; Martindale et al., 2013). The REE fractionation is imparted to the metasomatised mantle wedge and thus transferred to the generated magmas, without requiring the involvement of garnet in the residue of the peridotitic mantle sources. This mechanism also explains the lack of excess ^{230}Th (Fig. 11) in the Central Mediterranean ultrapotassic mafic rocks, which would be expected if the origin involved the melting of a garnet-rich mantle source (Avanzinelli et al., 2008). On the other hand, the lower Th/Nb and Th/U ratios, coupled with high HFSE concentrations (Fig. 11) and the significant excess ^{238}U (Fig. 10) observed in young post-leucititic *Roman* rocks (Avanzinelli et al., 2008) point to the major role of a recent fluid-like metasomatic component derived from allanite/monazite-saturated subducted sediment.

The SaLaThO geochemical paradox

The leucite-free lamproitic rocks of the *Tuscany and Corsican magmatic provinces* are part of a wider group of subduction-related potassic lamprophyric to lamproitic rocks. They are distributed along orogenic belts related to Tethys closure (e.g., Tibet, Turkey, Serbia, Macedonia, Western Alps, and Spain), also known as the Tethyan Realm Lamproites (Tommasini et al., 2011). These rocks are characterised by a positive correlation between Th/La and Sm/La (Fig. 12), which is opposite of that observed in typical arc-related magmas (Plank, 2005). This anomalous positive array points to a possible geochemical reservoir with high Sm/La and extraordinary high Th/La ratios named 'SaLaThO' by Tommasini et al. (2011). In figure 12 the mafic high-MgO potassic and ultrapotassic rocks of the Central Mediterranean region show two-fold behaviour. Lamproites and shoshonites of the *Corsica* and *Tuscany magmatic provinces* overlap with the positive array of Tethyan Lamproites, which ideally bridges the Th/La and Sm/La ratios of upper crustal rocks and sediments, and those of the SaLaThO component (Fig. 12). Rocks from the *Roman magmatic province* show the opposite trend with a less pronounced increase in Th/La (Fig. 12) and a negative correlation with Sm/La, as expected for typical subduction-related magmas (Plank, 2005).

The anomalous positive correlation between Th/La and Sm/La values observed in the early leucite-free ultrapotassic rocks (lamproites to shoshonites) has been related to a double metasomatic process by Tommasini et al. (2011). This involves recycled sediments (i.e., high Th/La and low Sm/La: Plank, 2005) accompanied by variable amounts, of a different metasomatic component characterised by high Sm/La and extremely high Th and Th/La ratio (i.e., SaLaThO). Ordinary metasomatic agents released during subduction, saturated in either allanite or monazite, are unable to produce a mantle source that would account for magmas with Th/La > 1, Sm/La > 0.3 and Th/U > 4. Tommasini et al. (2011) suggested that the source of magmas with such high Th/La and Sm/La requires the addition of a peculiar upper-crust-derived enriching agent to a depleted subcontinental lithospheric mantle. Stabilisation of lawsonite and zoisite/epidote at high P and low T within the subducted oceanic crust is the most likely process responsible for the SaLaThO geochemical component. These minerals are the major repository of Sr, Pb, U, Th, and LREE in such rocks (e.g., Spandler et al., 2003; Usui et al., 2006; Martin et al., 2014, and references therein). The lack of the SaLaThO geochemical component in the mafic high-MgO ultrapotassic rocks of the *Roman magmatic province* is a clear evidence for the arrival in the Central Mediterranean subcontinental lithospheric mantle of a new crustal-derived K-rich metasomatic agent after SaLaThO exhaustion. This new enriching agent is similar in nature and has similar trace element distribution to the *Corsica* and *Tuscany* leucite-free lamproites, although with lower Th/La ratios (Fig. 12).

In summary, a subcontinental lithospheric mantle capable of producing magmas with a SaLaThO signature is thought to be the result of anomalous enrichment caused by two distinct metasomatic agents. The first derived from partial melting of recycled sediments within the mantle wedge via subduction, and the second one, featuring high

Sm/La and Th/La ratios, derived by dehydration of the subducted and altered oceanic crust, in which re-mobilisation of lawsonite and epidote occurred (Tommasini et al., 2011). Such a geodynamic setting is likely to have been characterised by a mature subcontinental mantle wedge that experienced multiple collisional events that produced mélange slices of oceanic crust and metasedimentary rocks just prior to complete subduction of the oceanic crust. The shift from leucite-free to leucite-bearing rocks (i.e., from the *Tuscany event* to the *Roman* event) is characterised by the disappearance of the SaLaThO component in favour of the sediment-derived one, possibly due to the SaLaThO component becoming exhausted with magma formation.

Shift from leucite-free to leucite-bearing ultrapotassic rocks

The shift from silica-saturated to silica-undersaturated (i.e., from leucite-free to leucite-bearing) ultrapotassic magmas with time is not only limited to a change in the compositional characteristics controlling the final mineral assemblages in volcanic rocks. It is also related to a strong increase in the rate of magma production and its subsequent release to the surface. Indeed, lamproites of the *Corsica* and *Tuscany magmatic provinces* are strongly volumetrically subordinate to shoshonitic mafic rocks and their derivative magmas. In contrast, in the *Roman magmatic province* leucite-bearing ultrapotassic magmas (leucitites and plagio-leucitites) dominate volumetrically over post-leucitite rocks, which are confined to the final stage of activity and do not occur in all the volcanic districts. Neapolitan volcanoes exhibit igneous activity younger than 300 ka, when post-leucititic magmas become dominant, with the notable exception of Vesuvius.

The low production rate of silica-saturated lamproitic magmas (i.e., leucite-free; Fig. 5A) was possibly related to the refractory nature of the subcontinental lithospheric component of the mantle wedge prior to metasomatism. Metasomatism would have partially re-fertilised the refractory peridotite to produce a phlogopite-bearing harzburgite to pyroxenite vein network, which then underwent partial melting with an excess of H_2O and low X_{CO2} (e.g., Foley and Venturelli, 1989). Melting experiments have shown that silica-rich lamproitic magmas are generated by partial melting of a lithospheric mantle, originally depleted in the basaltic component (e.g., Edgar, 1987; Foley, 1994; Foley et al., 2009) and with modal phlogopite stabilised, at the expense of olivine, by the uprising melts derived from partial melting of recycled sediment (e.g., Wyllie & Sekine, 1982; Sekine & Wyllie, 1983; Mallick et al., 2015). Conversely, silica-undersaturated kamafugitic to leucititic ultrapotassic magmas (kalsilite- and leucite-bearing) are generated in a metasomatised upper mantle by partial melting of a phlogopite-bearing peridotite, controlled by a excess CO_2 under high X_{CO2} (e.g., Wendlandt and Eggler, 1980a; Thibault et al., 1992; Dasgupta and Hirschmann, 2006). Considering that the mantle beneath the Central Mediterranean region is particularly depleted in the basaltic component (prior to metasomatism), as illustrated by the major and trace element contents of the most primitive ultrapotassic magmas (e.g., lamproites and kamafugites), re-fertilisation is required to produce a sublithospheric mantle source containing modal clinopyroxene and phlogopite in place of orthopyroxene and phlogopite (e.g., Edgar, 1987).

In continental itra-plate settings the CaO re-fertilisation of subcontinental lithospheric mantle under excess CO_2 to generate ultrapotassic magmas may be achieved through carbonatitic metasomatism. The association of low-silica and high-potassium kamafugites with carbonatites in Central Africa is an example of coupling high-K_2O and carbonate-rich conditions during metasomatism (e.g., Lloyd et al., 2002; Foley et al., 2009).

Kamafugites also occur in the Central Mediterranean region kamafugites. The possible synchronous occurrence of carbonatite with kamafugite is still matter of debate (e.g., Stoppa and Wolley, 1997; Peccerillo, 1998), although it has been experimentally shown that Umbrian kamafugites might be the silicate counterparts of an unmixing process with carbonatitic melts (Martin et al., 2012). However, trace element distributions in Central Mediterranean kamafugites (Fig. 6C) show strong differences in terms of Th, Pb, REE, and HFSE when compared with the intraplate continental kamafugites, leucitites, and carbonatites (Fig. 7D), while still remaining similarities with the lamproitic to shoshonitic and calc-alkaline igneous rocks of the *Corsica* and *Tuscany magmatic provinces* (Figs. 7A and 7B). According to the experimentally determined partition coefficients (e.g., Dasgupta et al., 2009; Martin et al., 2012), the peaks at Pb and Sr, along with the troughs at Nb, Ta, and Ti, cannot be explained by simple element partitioning between immiscible carbonatite and silicate melts or by primary carbonatite formation in peridotitic mantles. In addition, Central Mediterranean kamafugites and associated leucitites and plagio-leucitites, show high Th/Nb and Th/U, and low Th/Rb ratios that overlap with the composition of Western Mediterranean lamproites (Fig. 10); these ratios are clearly different from those of continental intraplate kamafugites and carbonatites (e.g., Rogers et al., 1992; Zhao et al., 2005; Eby et al., 2009; Rosenthal et al., 2009; Brod et al., 2013). In addition, the radiogenic isotopic compositions of the Central Mediterranean kalsilite- and leucite-bearing igneous rocks are plotted toward a clear crustal component (Figs. 8 and 9) rather than overlapping with typical mantle isotope compositions (similar to the so-called FoZo), in contrast to continental intraplate carbonatites, kamafugites and leucitites (e.g., Carlson et al., 2007; Guarino et al., 2013).

To explain the peculiar crustal-like geochemical characteristics of kalsilite- to leucite-bearing Central Mediterranean ultrapotassic rocks, we infer a mantle source metasomatised by a carbonate-rich and potassium-rich component derived from carbonate-rich metapelites recycled via subduction within the mantle wedge. Partial melting of recycled carbonated pelites to produce K-rich carbonatites that generate mantle metasomatism has been shown to occur at depth within the mantle wedge (e.g., Thomsen & Schmidt, 2008; Poli et al., 2009; Grassi and Schmidt, 2011a, b). The reaction of these sediment-derived K-rich carbonatitic melts with depleted peridotite acts to re-fertilise the subcontinental upper mantle. It stabilises diopside and forsterite at the expense of enstatite and carbonated melt, in addition to phlogopite and phengite as K-repository phases (e.g., Yaxley et al., 1991, 1998). Partial melting of such metasomatised veins at relatively low pressure and high X_{CO2} produces strongly silica-undersaturated ultrapotassic magmas, from kamafugitic to leucititic in composition (e.g., Wendlandt & Eggler 1980a). Sedimentary carbonates were expected to behave as refractory phases at sub-arc depths of 180 km (Yaxley and Green 1994; Schmidt et al., 2004), but recent experimental studies contradict this view. Indeed, they have shown that carbonate-saturated pelites may produce potassic carbonatitic melts at relatively low temperatures (800-1000 °C) under a wide range of pressures (2.4-3.5 GPa; e.g., Thomsen and Schmidt, 2008; Poli et al., 2009; Grassi and Schmidt, 2011a, 2011b; Tumiati et al., 2013), inducing potassium- and carbonate-type metasomatism within the mantle wedge. Recent experimental studies have shown that carbonatitic liquids can form within this range of T° at shallow depths (ca. 120 km) at destructive plate margin by subduction of carbonate-rich sediments to form a carbonatitic metasomatic agent (Poli, 2015). Liu et al. (2015) has also recently reported evidence for sedimentary carbonate recycling from subduction-related xenoliths.

K-rich carbonatitic metasomatic melts from recycled marly sediment are extremely mobile and reactive and so are expected to percolate into the mantle wedge. Reaction of these melts with a depleted mantle is then able to produce CO2-rich phlogopite-bearing wehrlite. This has a suitable mineralogy to represent the source for kamafugitic to leucititic magmas (e.g., Arima and Edgar, 1983; Edgar, 1987; Conticelli, 1989; Lloyd et al., 1996).

The need for allanite and monazite in carbonate-rich metasomatic melts (see above) provides similar temperature constraints: in the pressure range 2.5-4.0 GPa the temperature required to exhaust allanite is >1000 °C (Hermann, 2002; Klimm et al., 2008). Given the doped composition of the experiments, which have shown residual monazite up to 900°C within a metapelitic system (Skora & Blundy, 2010), this value could be an over-estimate. Further experimental studies on trace element partitioning, during the melting of a carbonate metapelite indicate that metasomatic melts with geochemical signatures comparable to those observed in Central Mediterranean silica-undersaturated magmas are observed at higher pressures (8-13 GPa) during crustal recycling within the mantle wedge at depth of 170-300 km (Grassi and Schmidt, 2012a,b). The temperature range for sediment melting (800–1100°C; Mann and Schmidt, 2015) indicated by the geochemistry of the Central Mediterranean rocks is consistently higher than that estimated at the slab–mantle interface by thermal models of both 'cold' and 'warm' subduction. To explain this apparent inconsistency we suggest three possible scenarios: i) an increase in the temperature of the slab–mantle interface due to locking of the subduction zone following continental collision; ii) the physical incorporation of portions of the subducted sediment from the top of the slab into the mantle, either by imbrication or via diapirs (e.g., Klimm et al., 2008) and their melting in the hot central region of the mantle wedge (Avanzinelli et al., 2009); and iii) flushing of sediment overlying the slab with fluids liberated by dehydration of the subducted oceanic crust (Kerrick and Connolly, 2001).

On the basis of the above evidence the shift from leucite-free to leucite-bearing ultrapotassic rocks, in the Central Mediterranean region, may have be related to a change in the carbonate contents of the sediment recycled via subduction varying over time from pelitic to carbonate-pelitic sediment. The decoupling of Sr and Nd isotopes between *Corsica*/*Tuscany* and *Roman magmatic provinces* (Fig. 8A) provides a first evidence of this change in composition of the sediment recycled (Conticelli et al., 2002). Further indications are provided by the correlation between $^{87}Sr/^{86}Sr_i$ and each of Ba/Sr and Ce/Sr, where the data from *Roman* mafic rocks bridges the gap between the upper crust silicate-rich rocks field and that of the limestones (Fig. 13). Given that carbonate sediment is enriched in Sr the shift to low Ba/Sr and Ce/Sr ratios, passing from leucite-free to leucite-bearing is consistent with the involvement of a carbonate-rich sedimentary component. A partial exception to this general trend is represented by the young Neapolitan rocks, including both Vesuvius plagio-leucitites and associated post-leucitites, which have low $^{87}Sr/^{86}Sr$ but show a clear shift toward high Ba/Sr. This feature is consistent with the recent addition of a further fluid-like component in the Neapolitan area, as previously discussed in order to explain the excess ^{238}U of the Neapolitan rocks (Avanzinelli et al., 2008).

Mineralogy and vein networks of the mantle source

Olivine and spinel are the first mineral phases to crystallise from mantle-derived magmas. Olivine is also the most abundant phase in the upper mantle and spinel is the most important repository for Al in peridotite from the lithospheric mantle. Therefore, the forsterite content [Fo = Mg/(Fe+Mg)] and the Cr# [Cr/(Al+Cr)] of olivine–spinel pairs that crystallize in primitive mafic magmas provide information about the fertile *vs.* refractory degree of the peridotitic components of their mantle source and the possible fractionated phases during the early stages of magma differentiation (e.g., Arai, 1994a,b; Prelević and Foley, 2007). The higher the Cr#, the more depleted the mantle source. Further information can be obtained from the contents of minor and trace compatible elements (e.g., Ni, Mn, Cr, and Co) in olivine phenocrysts that crystallised in the erupted mafic volcanic rocks. These contents depend on the modal abundance of olivine in the magma source (e.g., Sobolev et al., 2005, 2007; De Hoog et al., 2010), while incompatible trace elements provide clues to the nature of the metasomatic agent (e.g., Foley et al., 2011, 2013; Prelević et al., 2013).

Early phenocrysts trace the peridotitic component of the source

Olivine–spinel pairs in the Central Mediterranean ultrapotassic rocks display large ranges in the Cr# of spinel and the forsterite content of olivine (Fig. 14). Several fractionation trends are defined for each recognised volcanic suite (Tab. 1), with high-forsterite olivine–spinel pairs falling well within the olivine–spinel mantle array (OSMA; Fig. 14; Arai, 1994a). The occurrence of euhedral spinel enclosed in high-forsterite olivine phenocrysts from Torre Alfina and Orciatico *Tuscany* lamproites show the highest Cr# ever seen in magmatic rocks (Fig. 14). This provides grounds to argue for equilibration of the magma with a mantle source assemblage, extremely depleted in Al and relatively enriched in Cr (e.g., Arai et al., 1994a; Conticelli, 1998; Prelević and Foley, 2007). The same holds true for olivine-spinel pairs from *Tuscany* shoshonites although the fractionation trend follows divergent pathways (Fig. 14). Olivine-spinel pairs from mafic high-MgO leucitites and plagio-leucitites of the *Roman magmatic province* (Fig. 14) have slightly lower Fo and Cr#, respectively, in comparison to the olivine-spinel pairs of the *Tuscany magmatic province*, close to the compositions observed of pairs falling within the mantle array (OSMA) and those in Spanish and Serbian lamproites (Prelević and Foley, 2007). The compositional variation of the high-forsterite olivine-spinel pairs within the mantle array (OSMA) show clear spatial trends, dominated with the lowest Cr# in spinels being hosted in olivine phenocrysts from the Roccamonfina and the Neapolitan volcanoes (Fig. 14), in the southernmost region of the *Roman magmatic province*. These data indicate a decrease in the residual character of the peridotitic component of mantle magma sources southward, from Tuscany and Latium to the Neapolitan area (Fig. 1).

Ni and Ca in olivine phenocrysts from mafic ultrapotassic Central Mediterranean rocks also discriminate between different volcanic suites (Fig. 14). Extraordinarily high Ca is stored in olivine phenocrysts from kamafugites and leucitites (Boari and Conticelli, 2007), while much lower contents are partitioned into olivine phenocrysts from lamproite and shoshonites of the *Tuscany magmatic province* (Conticelli et al., 2013). Kirschsteinite to monticellite are observed in the most silica-undersaturated ultrapotassic rocks (e.g., Boari and Conticelli, 2007; Melluso et al., 2010). The reverse is true for Ni, which occurs in high levels in olivine phenocrysts from mafic leucite-free ultrapotassic rocks (e.g., lamproite and shoshonites of the *Tuscany magmatic province*; Conticelli et al., 2013), whit the lowermost levels being observed in kalsilite- and leucite-bearing ultrapotassic rocks (Fig. 14). Prelević et al. (2013) suggested that the very high Ni contents of high-forsterite olivine in lamproites are related to an olivine-free mineralogy in the mantle source of lamproitic magmas from the Western Mediterranean. Indeed, because Ni is the most compatible trace element in olivine, the extraordinarily high Ni concentrations in magmatic olivine is only possible if the mantle source is not saturated in this mineral, indicating a pyroxenitic mantle source and a low bulk partition coefficient for Ni (e.g., Sobolev et al., 2005, 2007; De Hoog et al., 2010). Ammannati et al. (2015) reported extremely low Ni in high-forsterite liquidus olivine phenocrysts from leucitites and plagio-leucitites of the Ro*man magmatic province* (Fig. 14). Using the same line of reasoning, the authors related the low Ni to an increase in the bulk partition coefficient for Ni during mantle melting, arguing for a mantle source enriched in olivine. Considering also the extraordinary amount of Ca partitioned in olivine from kalisilite- to leucite-bearing rocks, a metasomatic reaction involving carbonate-rich metasomatic melts was proposed. Carbonatite-like metasomatic melts would then be able, at relatively low pressures

(< 2 GPa), to react with the peridotite in order to give a fine-grained olivine+clinopyroxene±chromite assemblage (e.g., Yaxley et al., 1991, 1998; Neumann et al., 2002). This possibility reinforces the hypothesis that the shift from the leucite-free lamproite-like magmas of the *Corsica* and *Tuscany magmatic provinces* to the leucite-bearing magmas of the *Roman magmatic province* is driven by carbonatitic potassium-rich metasomatic melts, derived from subducted recycled carbonated metapelities and transferred into the mantle wedge.

The smoking gun: Lithium in olivine phenocrysts

Lithium is a moderately incompatible trace element with respect to the mineralogy of the bulk mantle. Due to its volatile character, it is enriched in the upper continental crust (up to 70 ppm in clastic sediment) compared with the lithospheric upper mantle (< 3 ppm; Seitz and Woodland, 2000). Olivine is the main Li reservoir in the mantle, although the content in olivine never exceeds 5 ppm (De Hoog et al., 2010). Higher Li concentrations in olivine (> 10 ppm, Neumann, 2002; Foley et al., 2013) are ascribed to the storage of recycled upper crustal sediment within the mantle; consequently, olivine can be safely used to trace the nature of mantle metasomatism (Prelević et al., 2013). Ammannati et al. (2015) reported the strong enrichment of Li in olivine phenocrysts in equilibrium with both mafic leucite-free and leucite -bearing ultrapotassic rocks, with values up to 35 and 50 ppm, respectively. In liquidus high-Fo olivine, Li concentrations of >5 ppm indicate that the olivine crystallised from a melt in equilibrium with a peridotitic source enriched in recycled Li from silicic sediment (either metapelitic or carbonate-rich), with no need to invoke the late-stage acquisition of Li during magma storage and ascent through the upper continental crust (Ammannati et al., 2015).

Vein networks and increasing vein-host rock interaction during melting

The recycling of sediment within the upper mantle occurs via partial melting, producing felsic melts that interact with peridotite during ascent; thus, the metasomatic mineralogy remains confined within a discrete peridotite-modified vein network (Foley, 1992b). Indeed, potassium-, silica-, and alumina-rich melts flowing through lithospheric peridotite react with olivine to produce K-bearing phases (e.g., phlogopite and K-amphibole), orthopyroxene, and minor clinopyroxene, leaving most of the surrounding mantle unaffected by this reaction (e.g., Wendlandt and Eggler, 1980b; Wyllie and Sekine, 1983; Foley, 1992b; Bianchini et al., 2011). To explain the SaLaThO geochemical components, Tommasini et al. (2011) suggested a more complex mechanism for the metasomatic enrichment of the mantle-source of the Mediterranean lamproitic magmas involving multiple metasomatic events. This indicates the existence of an older set of veinlets that had accommodated the SaLaThO geochemical component within the subcontinental lithospheric upper mantle.

Central Mediterranean ultrapotassic mafic rocks show a systematic decrease in potassium and related incompatible elements over time within each magmatic event (Fig. 15). This is the case for both the *Corsica* and *Tuscany magmatic provinces* (leucite-free ultrapotassic rocks; Fig. 5A) and the *Roman magmatic province* (leucite-bearing ultrapotassic rocks; Fig. 5A). The transition from ultrapotassic to shoshonitic and calc-alkaline volcanic suites is widely observed in all potassic associations throughout the Mediterranean basin (e.g., southeast Spain, the Western Alps, Serbia, Montenegro, and Turkey; e.g., Altherr et al., 2004; Duggen et al., 2005; Prelević et al., 2004, 2008, 2012, 2015; Peccerillo and Martinotti, 2006; Owen, 2008; Conticelli et al., 2009a). In some cases the correlation with time is unclear. However, the best evidence of temporal variations in geochemistry has been observed in the *Tuscany* and *Roman magmatic provinces*. In most cases the stratigraphy and geochronology related to a single plumbing system affirm a change from either lamproite- or kamafugite-like rocks, to shoshonites and high-K calc-alkaline rocks, through to shoshonites or leucitites (e.g., Perini et al., 2004; Conticelli et al., 2009a, 2009b, 2011, 2013; Frezzotti et al., 2007; Boari et al., 2009b). At Somma-Vesuvius volcano, however, the opposite is observed; plagio-leucititites follow trachybasaltic to trachytic volcanic rocks (e.g., Cioni et al., 2008, and references therein) in a similar succession to that observed at Stromboli volcano in the Aeolian Arc (e.g., Francalanci et al., 1989, 2004, 2007).

The geochemical transition between magmatic series with different levels of K and incompatible trace element enrichment (i.e., from ultrapotassic to shoshonites or vice versa) has been explained in at least three different ways, as follows: i) relaxation of the isotherm that intercepts the solidus of variably metasomatised upper mantle, with the most strongly metasomatised mantle found at depth (e.g., Peccerillo, 2005); ii) an increase or decrease in the assimilation of shallow-level carbonate with time, with the former being used to explain the Somma-Vesuvius case; and (Iacono-Marziano et al., 2007); iii) incremental partial melting of a mantle source metasomatised in a veined network and subsequent dilution of the vein end-member through interaction with surrounding peridotitic host rocks (e.g., Foley, 1992b; Conticelli et al., 2009a; Boari et al., 2009b). The first hypothesis fails to explain primitive mafic magmas from a single plumbing system, independent of their K_2O contents degree of silica-saturation, appear to have been equilibrated with a mantle source that is characterised by similar pre-metasomatic (Conticelli et al., 2007). Where the magma chamber has been confined within limestone, crustal assimilation of carbonate together with concurrent clinopyroxene fractionation (Iacono Marziano et al. (2007) can, in specific cases, be an effective process of differentiation (e.g., Peccerillo, 1998; Dallai et al., 2004; Boari et al., 2009a; Gaeta et al. 2009). The assimilation of limestone assimilation, however, can account for neither the decrease nor the increase of potassium and incompatible elements in primitive magmas that are in equilibrium with high-forsterite olivine (e.g., Savelli, 1967; Boari et al., 2009a). In the vein network model, the mantle source reacts differently during the upward migration of the isotherms, related to the tectonic and geodynamic evolution of a subduction system. Initially, partial melting affects the mantle portion with the lowest solidus temperature, evidence of which is found in the metasomatised mineralogy of the veins (Foley, 1992b). Any further temperature increase, due to post-orogenic isotherm re-equilibration, may also trigger partial melting of the surrounding mantle. This would allow metasomatic veins and host rock to interact, resulting in dilution of the metasomatised component.

Incompatible trace element and isotopic variations observed for the *Corsica*, *Tuscany*, and *Roman magmatic provinces* (Fig. 15), with the exception of the Neapolitan volcanoes, can be explained by mixing with a strongly alkaline component. This is either a lamproitic component for the *Corsica* and *Tuscany* leucite-free rocks, or a kamafugitic component for the *Roman* leucite-bearing ones, with a high-K calc-alkaline end-member. Dilution of the geochemical contribution from the veins occurs due to interaction with increasing proportions of the surrounding lithospheric mantle during partial melting. This produces isotopic mixing trends that are not distinguishable from those produced by simple mixing between two magmas. However, simple magma mixing at a shallow level within the upper crust has to be ruled out on petrographic grounds. Indeed, most of the high-MgO rocks have compositions that are very close to those of primary magma, whereby high-Fo olivine is in equilibrium with the bulk rock. If magma mixing occurred at shallow levels, olivine would have crystallised and been left behind, driving the composition of the magma away from equilibrium with olivine itself.

Geodynamic and petrologic evolution

On the basis of the petrological, geochemical, and isotopic data reported in this study, we have produced a model to account for the geodynamic and petrologic evolution of the Central Mediterranean area. This model is relevant to our understanding of the shift from silica-saturated to silica-undersaturated magmas from the Miocene through to the present time (Fig. 16).

Corsica magmatic event

During the Miocene, leucite-free ultrapotassic (lamproites) and associated igneous rocks (shoshonites to high-K calc-alkaline rocks) formed the *Corsica magmatic province*. They were emplaced along the Eastern edge of the Corsica–Sardinia continental block, both inland (Sisco, Corsica) and offshore (Capraia Island and the Cornacya submarine bank; Fig. 1). Subducted metapelites underwent partial melting (Fig. 16a) producing metasomatic melts with residual garnet and rutile. The reaction of these potassium- and silica-rich metasomatic melts with the subcontinental lithospheric mantle overprinted the pre-existing SaLaThO geochemical component (Fig. 12), forming a new phlogopite-bearing, orthopyroxene-rich and olivine-poor/absent vein network (see upper petrographic illustration in Fig. 16a).

Silica-saturated lamproitic magmas at Sisco (Corsica) were produced by small degrees of partial melting of metasomatic veins, which have the lowest solidus with respect to the surrounding unaltered mantle wedge (Foley, 1992b). The subducted slab rolled back, eastward, and the Apennine orogeny also moved eastward (anticlockwise) (Civetta et al., 1978; Channell, 1996; Faccenna et al., 2001, 2004; Savelli, 2002; Caricchi et al., 2014). At the beginning of the Eocene the area between Corsica and Tuscany underwent post-orogenic extension, with initial opening of the Tyrrhenian basin and uprising of the asthenospheric mantle (e.g., Malinverno and Ryan, 1986; Jolivet et al., 1996; Doglioni et al., 1996) and subsequently of associated geotherms. Later, incremental post-subduction re-adjustment of the isotherms, which followed post-orogeny extension, triggered partial melting. Initially this was only within the phlogopite-bearing pyroxenite of the vein (see the upper petrographic illustration in Fig. 16). However, after a further temperature increase, partial melting also started to involve the peridotite host rock, producing primary magmas less enriched in alkali and incompatible elements, but still retaining subduction-related signature (Figs 6, 8, and 9).

Tuscany magmatic event

From the lower Pliocene, leucite-free ultrapotassic (lamproites) and associated igneous rocks (shoshonites to high-K calc-alkaline rocks) concentrated on the western side of the Italian Peninsula formed the *Tuscany magmatic province*. These rocks occur as sub-volcanic bodies and small lava flows, located mainly inland (e.g., Orciatico, Montecatini val di Cecina, Temperino valley, Radicofani, and Torre Alfina) and rarely offshore (Elba Island dykes; Fig. 1). The subducted slab continued to supply metapelitic sediment at depth, which was recycled (Fig. 16B) and then reacted with the surrounding depleted lithosphere. This is a similar process to that suggested for the Corsica event, producing phlogopite/amphibole-bearing orthopyroxenitic veins (see the upper petrographic illustration in Fig. 16).

At the same time, the Tyrrhenian post-orogenic basin continued to open, with the initial crustal extension moving inland due to the Apennine orogeny, and a further eastward jump of the subducted slab (e.g., Malinverno and Ryan, 1986; Doglioni, 1991; Jolivet et al., 1996; Bonini and Sani, 2002). Similarly to Corsica, in Tuscany partial melting was triggered during post-orogenic extension following the eastward migration of the slab. Partial melting first occurred within the vein mineralogy because of its lower solidus temperature producing lamproite magma The lowest solidus temperature occurs at the intersection between the SaLaThO and Phl/Amp pyroxenitic veins (see the upper petrographic illustration in Fig. 16) and it produces silica-saturated lamproite-like magmas. A further temperature increase extended the partial melting to the host peridotite producing a large spectrum of primary-mantle-derived magmas with decreasing alkalinity and potassium content with time (e.g., Conticelli et al., 2007, 2009a, 2011). Shoshonitic to high-K calc-alkaline Tuscany primary magmas are less enriched in potassium and related incompatible elements (Fig. 15) but retain subduction-related signatures (Figs 6, 8, and 9).

Roman magmatic event

From the upper Pleistocene, (Ionian) kalsilite-(kamafugites) to leucite-bearing (leucitites and plagio-leucitites) igneous rocks of the classic *Roman magmatic province* were erupted along the main extensional graben bordering the the eastern side of the Apennine chain, from Monte Amiata to the Neapolitan area (Fig. 1). A time gap of ca. 200 ky separates the newly erupted leucite-bearing magmas of the *Roman magmatic province* from the final differentiated magmas of the previous *Tuscany magmatic province* (Fig. 2), and hybrid rocks occur at the boundary between the two events (i.e., Amiata, Bolsena, and Cimino/Vico volcanoes)(e.g., van Bergen et al., 1983; Cioni et al., 1987; Conticelli et al., 2013, 2015). The start of this period of magmatism was preceded by the arrival, via subduction, of carbonated metapelites at depth (Fig. 16C), which were partially melted to produce lime-, CO_2-, and potassium-rich (carbonatitic) metasomatic melts (e.g., Thomsen and Smidth, 2008; Grassi and Schmidt, 2011a, 2011b) that moved into the lithospheric mantle wedge. The newly formed silica-poor and K-rich carbonatitic melts reacted with subcontinental lithospheric mantle to produce a new mineral assemblage of phlogopite/amphibole, olivine, and clinopyroxene, all concentrated in a vein network (see the bottom petrographic illustration in Fig. 16). The newly formed metasomatic melts may have arrived when the pre-existing SaLaThO metasomatic component was completely exhausted by melting related to Tuscany magmatism. Partial melting of the vein assemblage produced strongly silica-undersaturated primitive magmas (i.e., kamafugites = kalsilite-bearing). The transition from kamafugites to leucitites and then to plagio-leucitites was time-related (Boari et al., 2009b) and this indicates a dilution of the alkaline component (K_2O and related incompatible elements) with an increasing aluminous character and an increasing degree of silica saturation (Fig 5A). Partial melting of the veins occurred incrementally, from veins changing to vein+wallrock and transitioning to plagio-leucitites, and possibly post-leucitites with time. Increased partial melting at higher temperatures diluted the carbonate component in favour of the ambient peridotitic component (Boari and Conticelli, 2007).

Towards the end of the middle Pleistocene (<400 ka), the subducted slab became near-vertical (Fig. 16D), which produced the present day geometry (Spakman and Wortel, 2004). The Apennine compression migrated farther eastward, causing the central portion of the Apennine chain to enter a new post-orogenic extensional regime (e.g., D'Agostino et al. 2009, 2011). Relaxation of geotherms triggered the formation of kamafugitic magmas that were emplaced in the central Appennines (e.g., Polino, San Venanzo, and Cupaello; Fig. 1), retaining geochemical signatures similar to those of other previous *Roman* kamafugites, leucitites, and plagio-leucitites (Figs 6–9).

Post-leucitites

Almost coeval with the occurrence of early kamafugites in the intrapennine region (Umbria), the volcanic plumbing of the pery-tyrrhenian *Roman* volcanic chain reached a magma production crisis. The new arrival of post-leucititic magmas (basalt to trachyte) in many *Roman* volcanoes triggered large parossistic eruptions that destroyed the stratovolcanoes. Volcanic activity was renewed following caldera formation, with the production in some cases of leucite-free magmas. *Post-leucititic* products reached the surface in these volcanic areas where leucititic magmas were completely exhausted and their magmatic reservoirs within the upper crust obliterated by caldera collapse (e.g., Conticelli et al., 2010). South of the Latium districts, post-leucitite magmatism shows distinctive characteristics indicating an origin from a less refractory peridotitic source (Fig. 14). This is reflected in a shift in the geochemical and isotopic composition of the magmas, indicating an asthenospheric mantle signature, either MORB-like (Fig. 8; e.g., Beccaluva et al., 1991; D'Antonio et al., 1999a) intraplate-like, possibly channelled by detachment of the Apennine–Calabria slab slab (e.g., Rosenbaum et al., 2008). In the Neapolitan area, U-series data indicate the recent arrival of slab-derived fluids that caused the resumption of magma production during the hystorical volcanic activity (e.g., Avanzinelli et al., 2008).

Acknowledgements

We deeply appreciate Tymothy Horscroft for inviting us to publish this paper on Lithos. Financial support has been provided by the 'Italian Ministry of Education and University' (MIUR) through PRIN 2010-2011 funds (grant 2010TT22SC_001). The authors wish to thank Massimo Mattei, Alan Edgar†, Giulia Perini, Rick Varne†, Elena Boari, Dejan Prelević, Rick Carlson, Claudio Faccenna, Leone Melluso, Tim Elliott, Simone Tommasini, Steve Foley, Angelo Peccerillo, Keith Bell, Giampiero Poli, Jon Davidson, and Fabrizio Innocenti for sharing ideas and discussions. Beatrice Bizzaro and Aaron Stallard are thanked for polishing the English language and style throughout the final version of the manuscript. The papers greatly benefitted by peer reviews made by anonymous and Michel Grégoire. Andrew Kerr is warmly thanked for his skilful editorial handling. The paper is dedicated to the memory of Anna Maria Farruggio.

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Figure Captions

Fig. 1 – Distribution of ultrapotassic rocks in the Central Mediterranean region; see text for further explanation. Legend: LMP = lamproites; Sho = ultrapotassic shoshonites, shoshonites and high-K calc-alkaline; Kam = kamafugites; Leu = leucitites and plagio-leucitites. Redrawn after Conticelli et al. (2010, 2013, 2015)..

Fig. 2 - Ages distribution in the ultrapotassic and associated rocks of the circum-tyrrhenian sector. For source of data see Marra et al. (2004), Laurenzi (2005), Peccerillo (2005), Conticelli et al. (2010).

Fig. 3 - Classification and Harker's diagrams for Italian potassic and ultrapotassic rocks: A) Total Alkali Silica diagram (Le Maitre, 2002); B) Potash vs. silica diagram for orogenic suites (Ewart, 1982); C_1) Mg-# vs. silica; C_2) Mg# vs. silica only for samples with Mg# > 60, with low porphyritic index, and olivine phenocrysts in equilibrium with bulk rock compositions. The complete set of data is available by download from the journal. Source of data: Rogers et al. (1985), Conticelli et al. (1987, 1991, 1992, 1997, 2001, 2002, 2009a, 2009b, 2011, 2013), Joron et al. (1987), Luhr and Giannetti (1987), Poli et al. (1987), Peccerillo et al. (1988), Conticelli and Peccerillo (1992), Orsi et al. (1995), Civetta et al. (1997), Ayuso et al. (1998), Conticelli (1998), di Battistini (1998), Bindi et al. (1999), D'Antonio et al. (1999a, 1999b), Pappalardo et al. (1999), Perini et al. (2000, 2004), Mascle et al. (2001), Gasperini et al. (2002), Avanzinelli et al. (2008), Prelević et al. (2008), Boari et al. (2007, 2009a, 2009b), Rouchon et al. (2008) and author's unpublished data.

Fig. 4 - Classification diagram for ultrapotassic rocks (Foley et al., 1987) with plotted the Italian ultrapotassic rocks selected to represent primitive mafic compositions. For source of data see figure 2.

Fig. 5 - A) Feldspathoid Silica-Saturation Index (FSSI) *vs*. Peralkaline Index (PeI) with plotted Central Mediterranean mafic potassic and ultrapotassic rocks (after Frost and Frost, 2008). Feldspathoid silica-saturation index calculated as normative *q*-[*lc*+2*(*ne*+*kp*)]. In this index normative *Ne* and *Kp* are multiplied by two because each mole of nepheline or kaliophilite consumes 2 moles of quartz to make silica-saturated feldspars (i.e., albite or orthoclase). FSSI > 0 the rocks are silica-oversaturated whilst FSSI < 0 the rocks are silica-undersaturated. Peralkaline Index (PeI) is defined as (K+Na)-Al on a molecular basis. Peralkaline rocks have PeI > 0, whereas from metaluminous and peraluminous rocks PeI < 0. B) K_2O/Na_2O *vs.* MgO wt.% used for discriminating between mafic potassic and ultrapotassic Central Mediterranean rocks. The lack of sample below 4 wt. % of MgO is due to the criteria used for sample selection (Fig. $3C_1$). Limits are drawn according to the criteria of (Foley et al., 1987). For source of data see figure 2.

Fig. 6 - Spiderdiagrams for mafic (Mg# > 0.60 and MgO > 4 wt.%) ultrapotassic and potassic igneous rocks of the *Corsica*, *Tuscany*, and *Roman* magmatic events (i.e., provinces), data are normalised to the primordial mantle values of McDonough and Sun (1995). When available, patterns for solid melt inclusions (MI) are plotted (black lines) (Nigokosian and van Bergen, 2010). Note that MI patterns are indistinguishable from those of the bulk rocks. Note also that the similarities in terms of troughs and peaks among patters of the different magmatic events piled up in the graph from the oldest (bottom) to the youngest (top) ones. Note also by comparison with figure 7 the similarities between the patterns of Italian ultrapotassic rocks and those of the Mediterranean sedimentary rocks (A and B), the slight differences in terms of Ba content with patterns of recent sediments from Sunda, Philippine, and Ande trenches and GLOSS. On the other hand, note the strong dissimilarities between patterns of the Italian ultrapotassic rocks and those of ultrapotassic rocks from within plate setting. For source of data see figure 2.

Fig. 7 - Spiderdiagrams for within plate ultrapotassic and carbonatitic igneous rocks, data are normalised to the primordial mantle values of McDonough and Sun (1995). Source of data are Fraser et al. (1985), Conticelli (1989, 1998), Davies and Lloyd (1989), Plank and Langmuir (1998), Graham et al. (1999), McDonald et al. (2001), Melluso et al. (2003, 2005a, 2005b), Boari et al. (2009b), Conticelli et al. (2009a), Eby et al. (2009), Rosenthal et al. (2009), and supplementary material of this paper.

Fig. 8 - Plots of $^{143}Nd/^{144}Nd_i$ (A) and $^{206}Pb/^{204}Pb_i$ (B) *vs.* $^{86}Sr/^{86}Sr_i$ for mafic (Mg-# > 0.60 and MgO > 4 wt.%) Central Mediterranean ultrapotassic and associated rocks; Subscript 'i' indicates that isotopic values were age corrected, thus calculated back at the initial value of the crystallisation of magma. Mantle zoo end members are from Stracke et al. (2005). The inset in A shows the isotope composition of solid melt inclusions from the potassic and ultrapotassic magmas object of this study (Korrnneeff et al. 2015), which are almost identical to the relative bulk host rocks and display the same overall variation of the whole dataset. Data are from Conticelli et al. (1992, 1997, 2001, 2002, 2007, 2009a, 2009b, 2011, 2013), Conticelli and Peccerillo (1992), Orsi et al. (1995), Civetta et al. (1997), Ayuso et al. (1998), Conticelli (1998), D'Antonio et al. (1999a, 1999b), Mascle et al. (2001), Pappalardo et al. (1999), Gasperini et al. (2002), Perini et al. (2003, 2004), Avanzinelli et al. (2008, 2012a, 2014), Prelević et al. (2008, 2010), Boari et al. (2009a, 2009b), supplementary material of this paper and author's unpublished data.

Fig. 9 - Plots of $^{176}Hf/^{177}Hf_i$ (A), $^{187}Os/^{188}Os_i$ (B), and $^{3}He/^{4}He$ (C) *vs.* $^{86}Sr/^{86}Sr_i$ for Central Mediterranean ultrapotassic and associated rocks; Subscript 'i' indicates that isotopic values were age corrected, thus calculated back at the initial value of the crystallisation of magma. The green curve in B represent mixing between a depleted mantle source with 3200 ppt of Os and $^{187}Os/^{188}Os$ = 0.13 (Walker et al., 1989) and a possible crustal component represented by an Italian crustal rocks reported in Conticelli et al. (2007) with 52 ppt Os and $^{187}Os/^{188}Os$ = 1.004. The red curve is the mixing curve between the least radiogenic sample (BR 29, see supplementary material) and the crustal rock as representative of a possible crustal contamination pattern. Data are from Gasperini et al. (2002), Conticelli et al. (2002, 2007, 2010), Martelli et al. (2004), Widom et al. (2004), Prelević et al. (2008, 2010, 2014), and Tommasini's unpublished data. Mantle Zoo end members are from Stracke et al. (2005).

Fig. 10 - ($^{230}Th/^{232}Th$) *vs.* ($^{238}U/^{232}Th$) equiline diagram for leucite-bearing rocks of the *Roman magmatic province* and younger leucite-free rocks of Neapolitan volcanoes (age < 300 ka). Parentheses indicate activity ratios. All activity ratios have been age-corrected. Data from Avanzinelli et al. (2008). The field for Etna is from Condominés et al. (1995).

Fig. 11 - Plots of Th/Rb, Th/Nb, and Th/U *vs.* Th for mafic (Mg-# > 0.60 and MgO > 4 wt.%) Central Mediterranean ultrapotassic and associated rocks; Fields of Western Alps (Conticelli et al., 2009a) and Spanish lamproites (Prelević et al., 2008; Conticelli et al. 2009a), on one end, and of Monticchio and Vulture volcanoes (Beccaluva et al., 2002; De Astis et al., 2006; author's unpublished data), and Monticchio carbonatite (D'Orazio et al., 2007)(*Lucanian magmatic province*), on the other end side, are also reported for comparison along with fields of continental within plate carbonatites and kamafugites from Central Africa (Davies and Lloyd, 1989; Graham et al., 1999; Eby et al., 2009; Rosenthal et al., 2009). For Central Mediterranean igneous rocks source of data see supplementary material of this paper, and author's unpublished data.

Fig. 12 - Plots of Th/La *vs.* Sm/La. Fields are drawn after: Arc Magmas from Plank (2005); Upper Crust from Melluso et al. (2003), Conticelli et al. (2009a), Mazzeo et al. (2014), and authors' unpublished data (supplementary

material); SaLaThO from Tommasini et al. (2011), G = Gloss is from Plank and Langmuir (1993, 1998). For Central Mediterranean igneous rocks source of data see supplementary material to this paper.

Fig. 13 - ($^{86}Sr/^{86}Sr)_I$ *vs.* Ba/Sr and Ce/Sr for mafic (Mg# > 0.60 and MgO > 4 wt.%) Central Mediterranean ultrapotassic and associated rocks, with reported the fields of within plate igneous rocks from the foreland (La Queglia, Pietre Nere, Pachino, Pantelleria, and Linosa; Avanzinelli et al., 2012b, 2014). G = Gloss (Plank and Langmuir, 1993, 1998); Upper Crust and limestones data are from Melluso et al. (2003), Conticelli et al. (2002, 2007, 2009a), Boari et al. (2009b), Mazzeo et al. (2014), and authors' unpublished data (supplementary material).

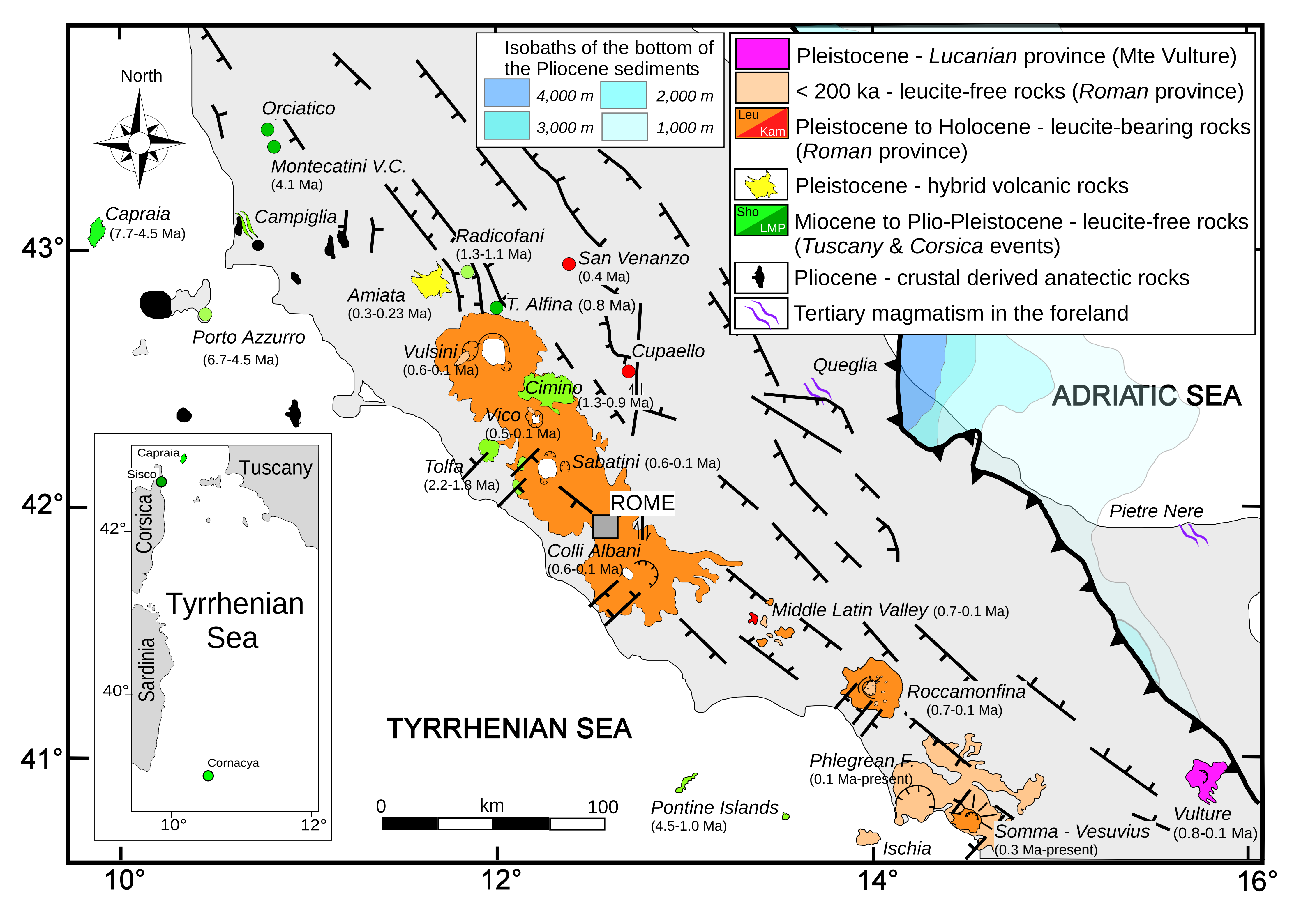
Fig. 14 - $[Cr/(Cr+Al)]_{spinel}$ (Cr-#), $Ca_{olivine}$, and $Ni_{olivine}$ *vs.* Forsterite $(Fo)_{olivine}$ for early euhedral phenocrysts in equilibrium with bulk mafic rocks (Mg# > 0.60 and MgO > 4 wt.%) of Central Mediterranean. Olivine Spinel Mantle Array (OSMA) is from Arai (1995a, b), data are from Conticelli et al. (1992, 1997, 2001, 2010b, 2011, 2013, 2014), Conticelli (1989, 1998), Perini and Conticelli (2002), Boari and Conticelli (2007), Ammannati et al. (2015).

Fig. 15 - Selected incompatible elements *vs.* potash contents in mafic (Mg# > 0.60 and MgO > 4 wt.%) Central Mediterranean ultrapotassic rocks. Note that La, Rb, and Nb are taken as examples of incompatible elements with different geochemical behaviour. Decrease of incompatible elements and K_2O within each magmatic province (*Tuscany* and *Roman*) with time are taken as an argument to support increasing vein-surrounding ambient peridotite interaction according to uprise geotherms after slab roll-back. For Central Mediterranean igneous rocks source of data see supplementary material to this paper, and author's unpublished data.

Fig. 16 - Cartoon showing the evolution with time of the subduction beneath the Italian Peninsula in the Central Mediterranean region, from Miocene to Middle Pleistocene. The geodynamic features are not to scale. The blow up depicts possible mineralogical assemblages of the veins and of surrounding lithospheric peridotitic mantle. Legend: OPX = orthopyroxene, PHL = phlogopite, AMPH = amphibole, OL = olivine, CPX = clinopyroxene, SP = spinel, TS = Tyrrhenian Sea. Drawn after Faccenna et al. (2001), Avanzinelli et al. (2009), and Ammannati et al. (2015). For further explanations see the text.

Table 1. Mineralogical and chemical characteristics of ultrapotassic and associate rocks from Central Mediterranean.

Epochs	Magmatic Province	Petrographic clan	K_2O/Na_2O	Rock type	Chemistry	Mineralogy	Note
Miocene (14 Ma) Plio-Pleistocene (5-0.9 Ma)	Corsica Tuscany	Lamproite	Ultrapotassic	Lamproite Orendite Minette Olivine-latite	CaO < 10 wt.% Na_2O < 1.5 wt.% Al_2O_3 < 12 wt.%	olivine (Fo_{93-59}), clinopyroxene, phlogopite, K-richterite, chromite, sanidine, picroilmenite, magnetite, apatite, thorite	plagioclase-free leucite-free silica-saturated *hypersthene normative*
Pliocene (11-7 Ma) Plio-Pleistocene (4-0.8 Ma)	Corsica Tuscany	Shoshonite	Potassic/ Ultrapotassic	Shoshonite Olivine-latite Latite Trachyte	CaO > 15 wt.% Al_2O_3 > 15 wt.%	olivine (Fo_{93-53}), chromite, clinopyroxene, plagioclase, sanidine, biotite, ilmenite, magnetite, apatite	plagioclase-free leucite-free silica-saturated *hypersthene normative*
Pliocene (11-7 Ma) Plio-Pleistocene (4-0.8 Ma)	Corsica Tuscany	High-K calc-alkaline	Sub-alkaline	K-andesite K-dacite K-rhyolite	CaO > 15 wt.% Al_2O_3 > 15 wt.%	olivine (Fo_{92-50}), chromite, clinopyroxene, plagioclase, sanidine, biotite, ilmenite, magnetite, apatite	plagioclase-free leucite-free silica-saturated *hypersthene normative*
Pleistocene (0.7 – 0.4 Ma)	Roman	Kamafugite	Ultrapotassic	Kalsilitite Kalsilit-melilitite	CaO > 15 wt.% Al_2O_3 < 14 wt.%	olivine (Fo_{93-31}), monticellite, clinopyroxene, phlogopite, chromite, melilite, leucite, kalsilite, perovskite, ilmenite, magnetite, apatite	feldspar-free kalsilite-bearing silica undersaturated *larnite normative*
Pleistocene (0.6 – 0.2 Ma) Holocene (Vesuvius)	Roman	Leucitite Plagio-leucitite	Ultrapotassic	Leucitite Basanite/tephrite Tephritic-phonolite Pnolitic-tephrite Phonolite	CaO > 15 wt.% Al_2O_3 > 14 wt.%	olivine (Fo_{91-24}), chromite, clinopyroxene, leucite, plagioclase, nepheline, phlogopite, melilite, magnetite, sphene, apatite	leucite-bearing silica undersaturated *leucite normative*
Pleistocene (0.2-0.1 Ma) Holocene (< 0.02 Ma)	Roman	Post-leucitite	Ultrapotassic	Trachybasalt Trachyandesite Latite Trachyte	CaO < 15 wt.% Al_2O_3 > 14 wt.%	olivine (Fo_{89-38}), chromite, clinopyroxene, plagioclase, nepheline, magnetite, apatite	Leucite-free silica saturated *nepheline normative*



Isobaths of the bottom of the Pliocene sediments
4,000 m
2,000 m
3,000 m
1,000 m
Pleistocene - Lucanian province (Mte Vulture)
< 200 ka - leucite-free rocks (Roman province)
Leu
Kam
Pleistocene to Holocene - leucite-bearing rocks (Roman province)
Pleistocene - hybrid volcanic rocks
Sho
LMP
Miocene to Plio-Pleistocene - leucite-free rocks (Tuscany & Corsica events)
Pliocene - crustal derived anatectic rocks
Tertiary magmatism in the foreland
North
Orciatico
Montecatini V.C.
(4.1 Ma)
Capraia
(7.7-4.5 Ma)
Campiglia
Radicofani
(1.3-1.1 Ma)
San Venanzo
(0.4 Ma)
Amiata
(0.3-0.23 Ma)
T. Alfina (0.8 Ma)
Porto Azzurro
(6.7-4.5 Ma)
Vulsini
(0.6-0.1 Ma)
Cupaello
Queglia
Cimino
(1.3-0.9 Ma)
ADRIATIC SEA
Vico
(0.5-0.1 Ma)
Tolfa
(2.2-1.8 Ma)
Sabatini (0.6-0.1 Ma)
ROME
Pietre Nere
Colli Albani
(0.6-0.1 Ma)
Middle Latin Valley (0.7-0.1 Ma)
Roccamonfina
(0.7-0.1 Ma)
TYRRHENIAN SEA
Phlegrean F.
(0.1 Ma-present)
Pontine Islands
(4.5-1.0 Ma)
Vulture
(0.8-0.1 Ma)
Somma - Vesuvius
(0.3 Ma-present)
Ischia
0
km
100
Capraia
Sisco
Tuscany
Corsica
Tyrrhenian Sea
Sardinia
Cornacya
42°
40°
10°
12°
43°
42°
41°
10°
12°
14°
16°

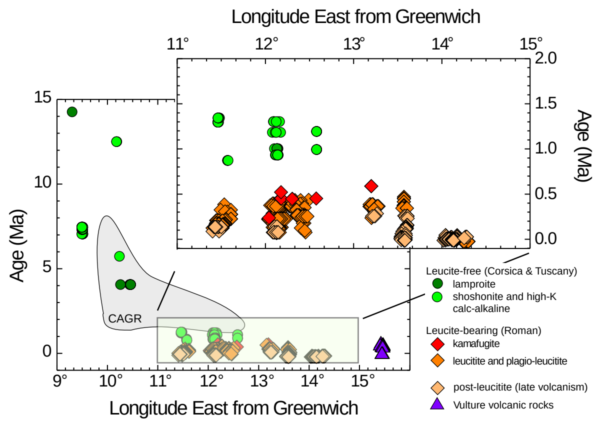


Figure 2 - Lithos - Conticelli et aal. (2014)
File: Lithos_Conticelli_FIG-02.CNV

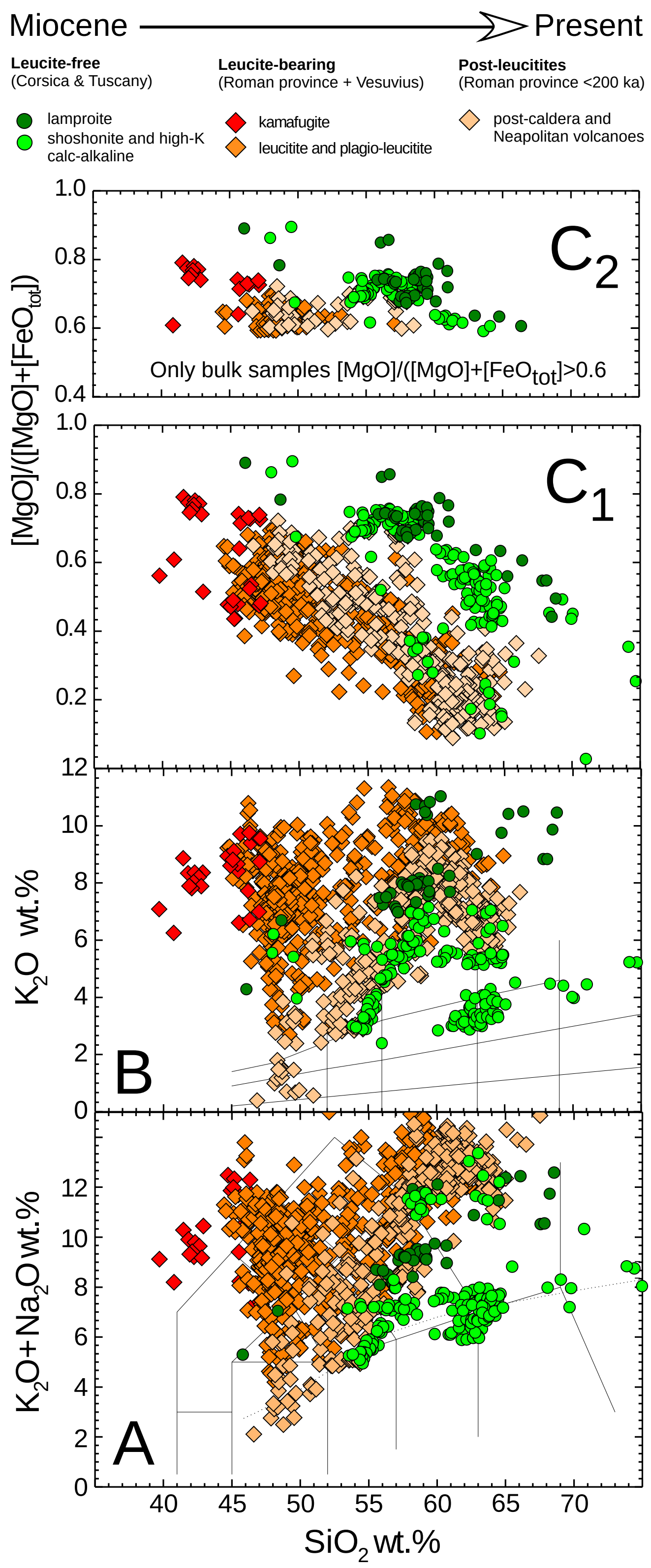


Figure 3 - Lithos - Conticelli et aal. (2014)
File: Lithos_Conticelli_FIG-03.CNV

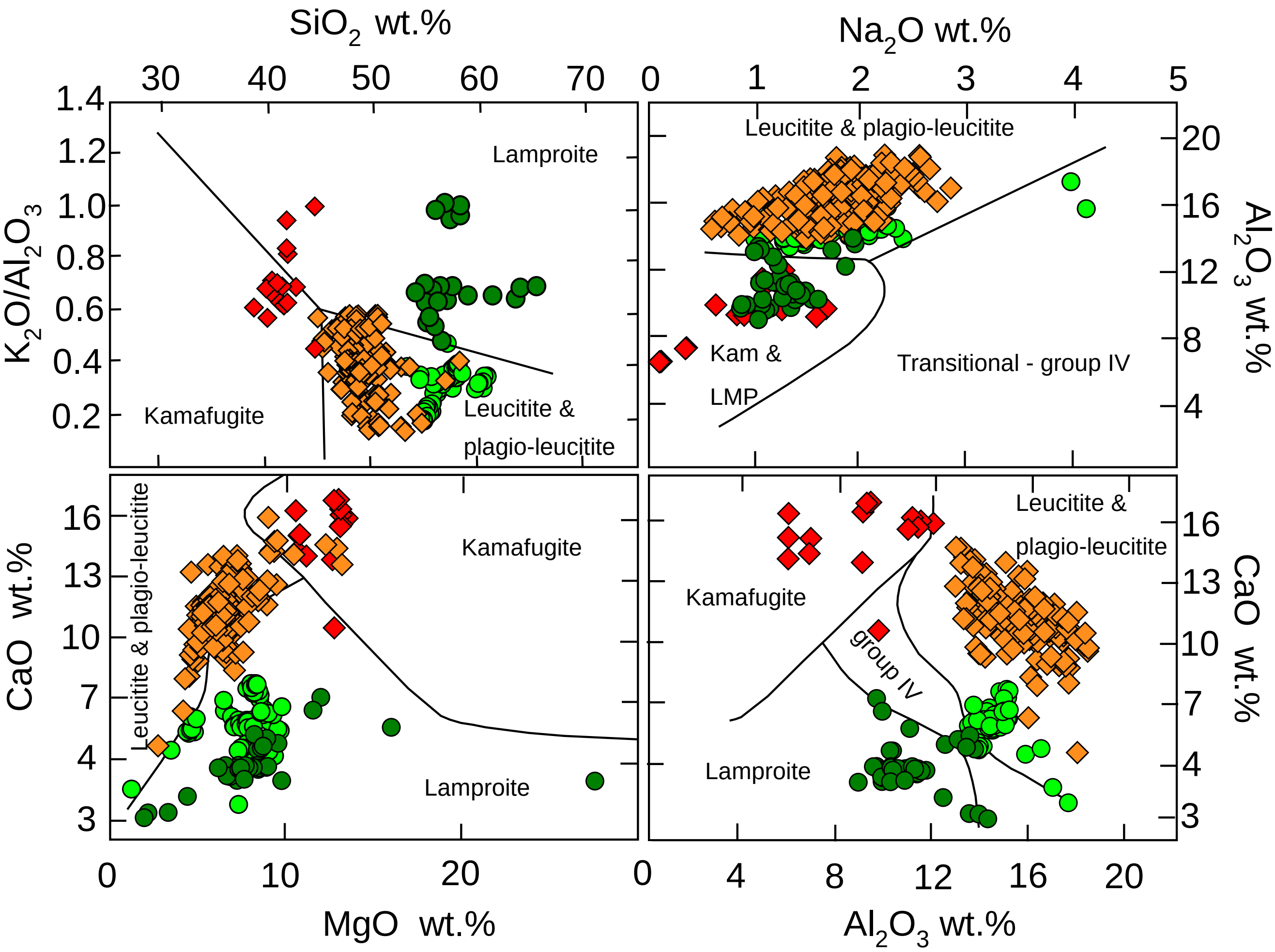


Figure 4 - Lithos - Conticelli et aal. (2014)
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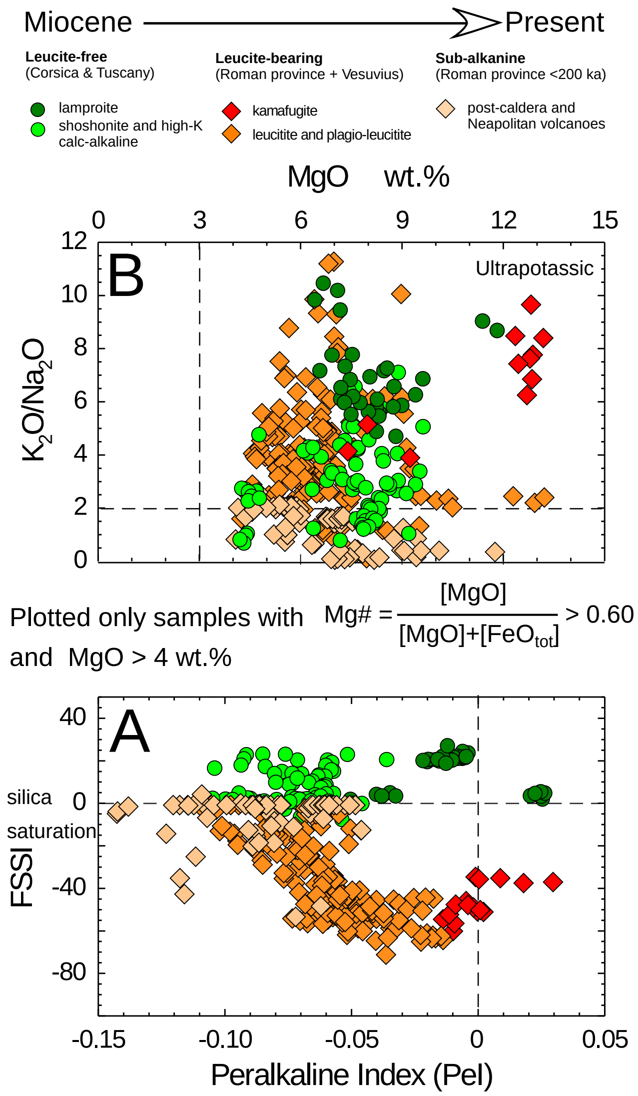

Figure 5 - Lithos - Conticelli et aal. (2014)
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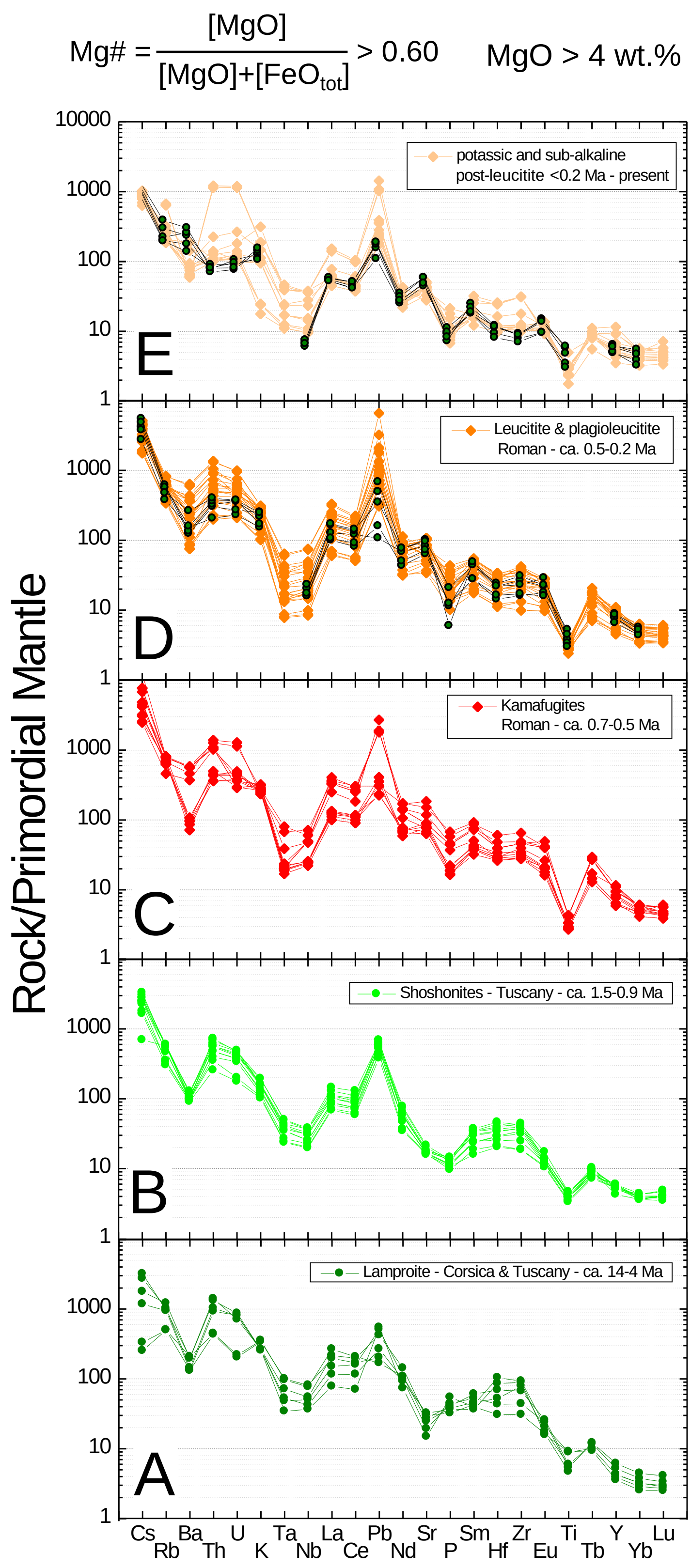

Figure 6 - Lithos - Conticelli et aal. (2014)
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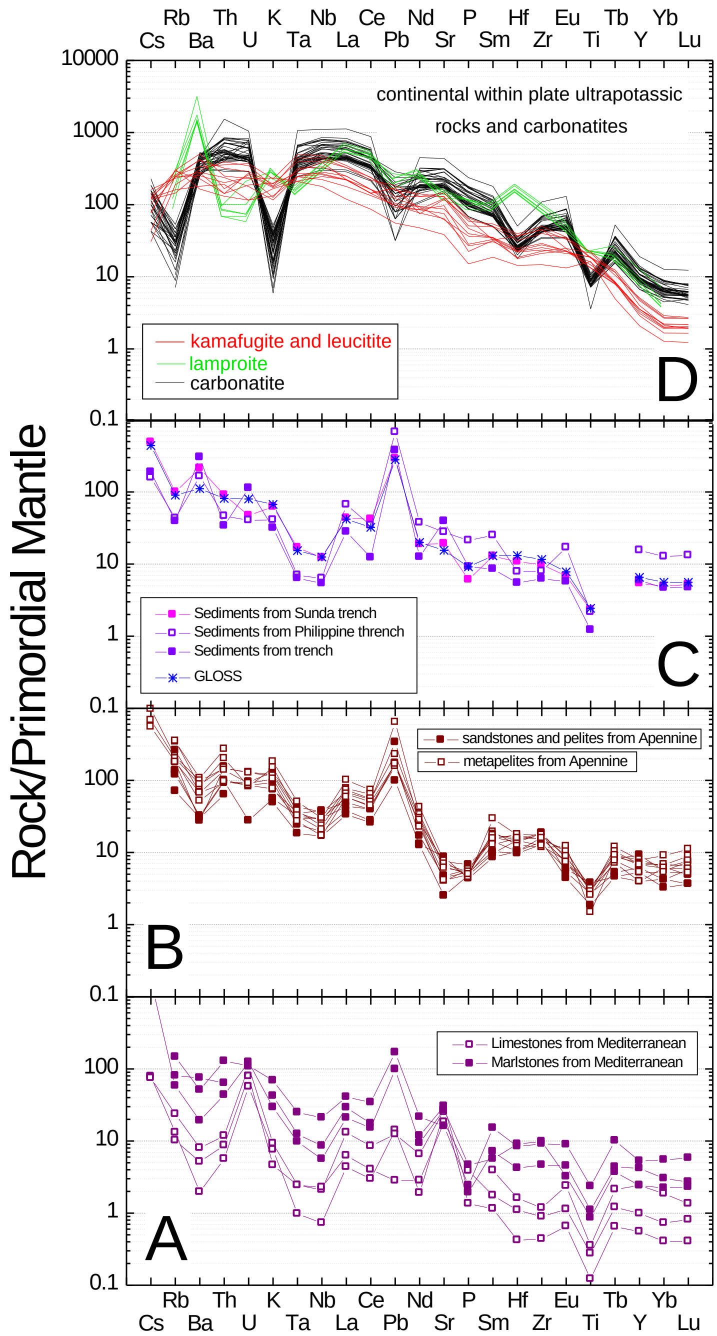

Figure 7 - Lithos - Conticelli et aal. (2014)
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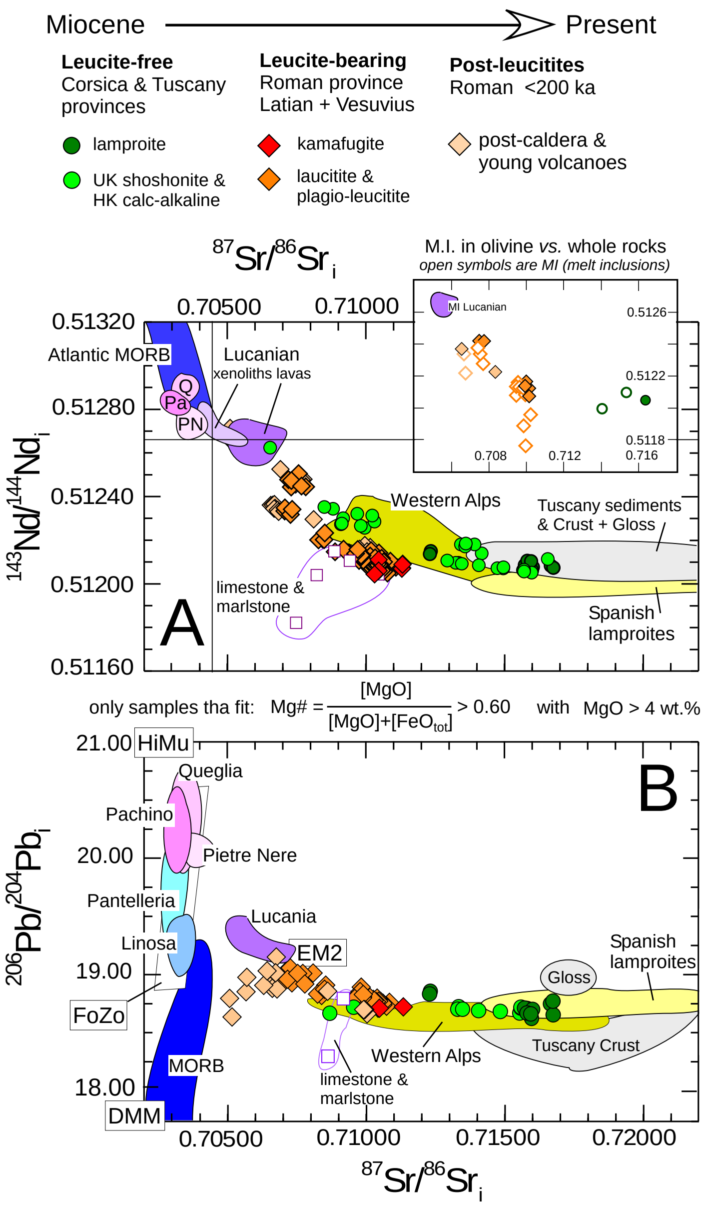

Figure 8 - Lithos - Conticelli et aal. (2014)
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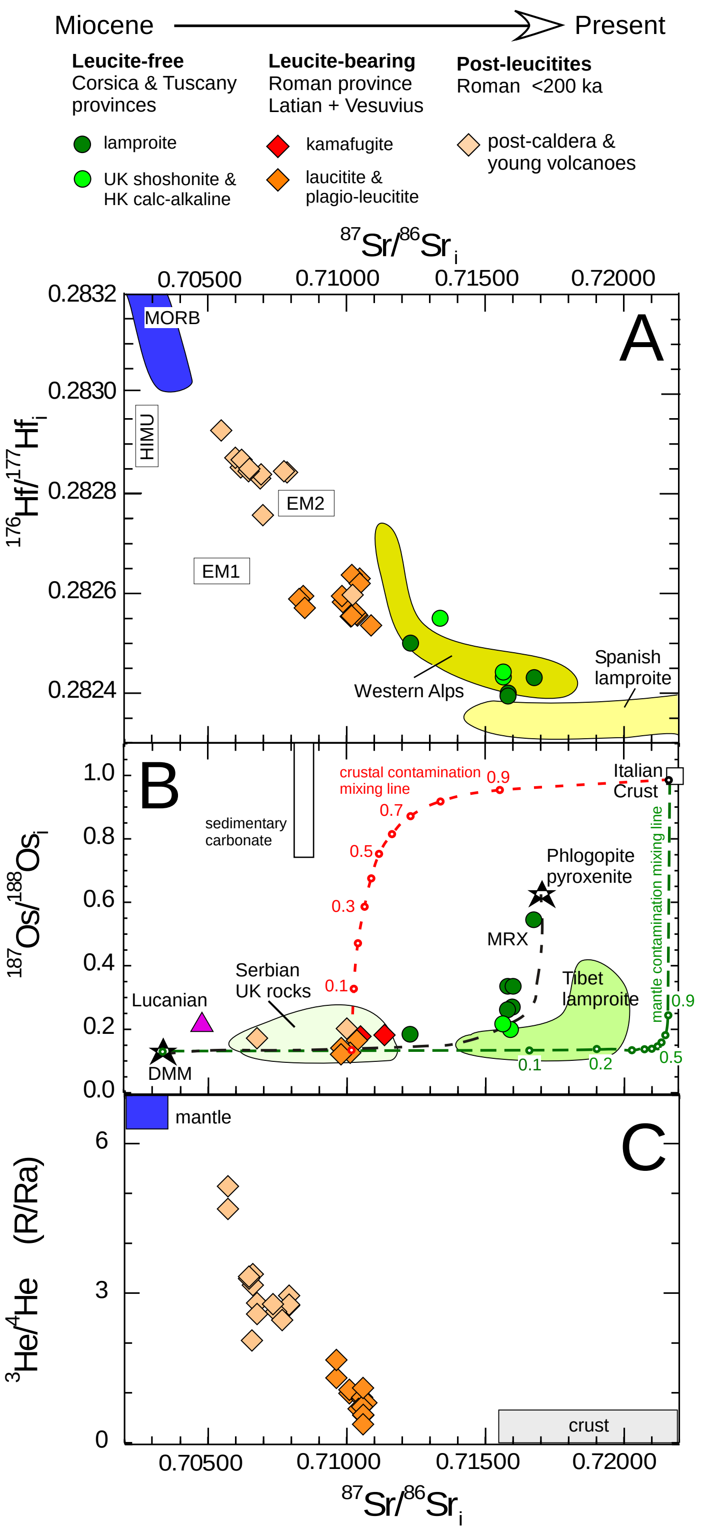

Figure 9 - Lithos - Conticelli et aal. (2014)
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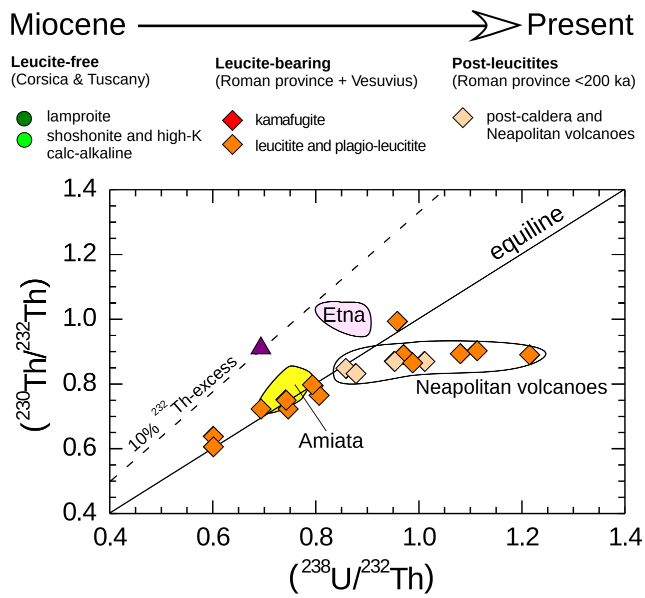

Figure 10 - Lithos - Conticelli et aal. (2014)
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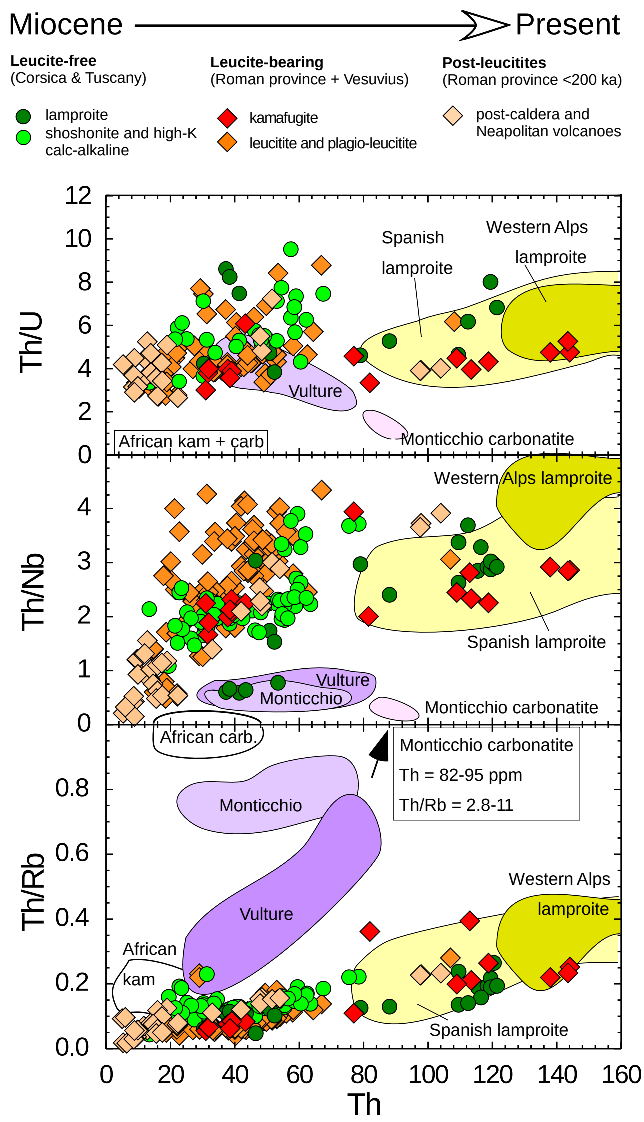

Figure 11 - Lithos - Conticelli et aal. (2014)
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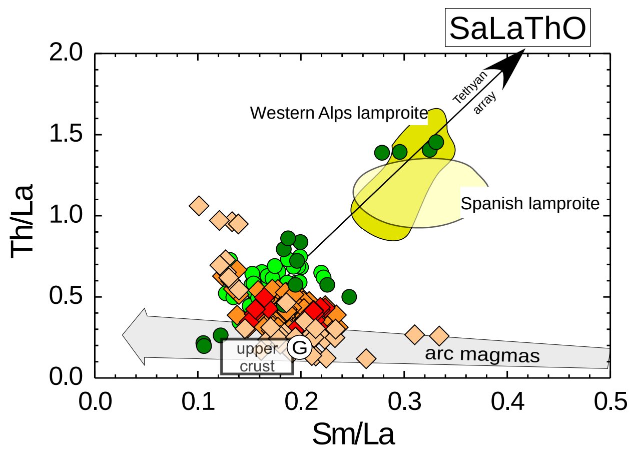

Figure 12 - Lithos - Conticelli et aal. (2014)
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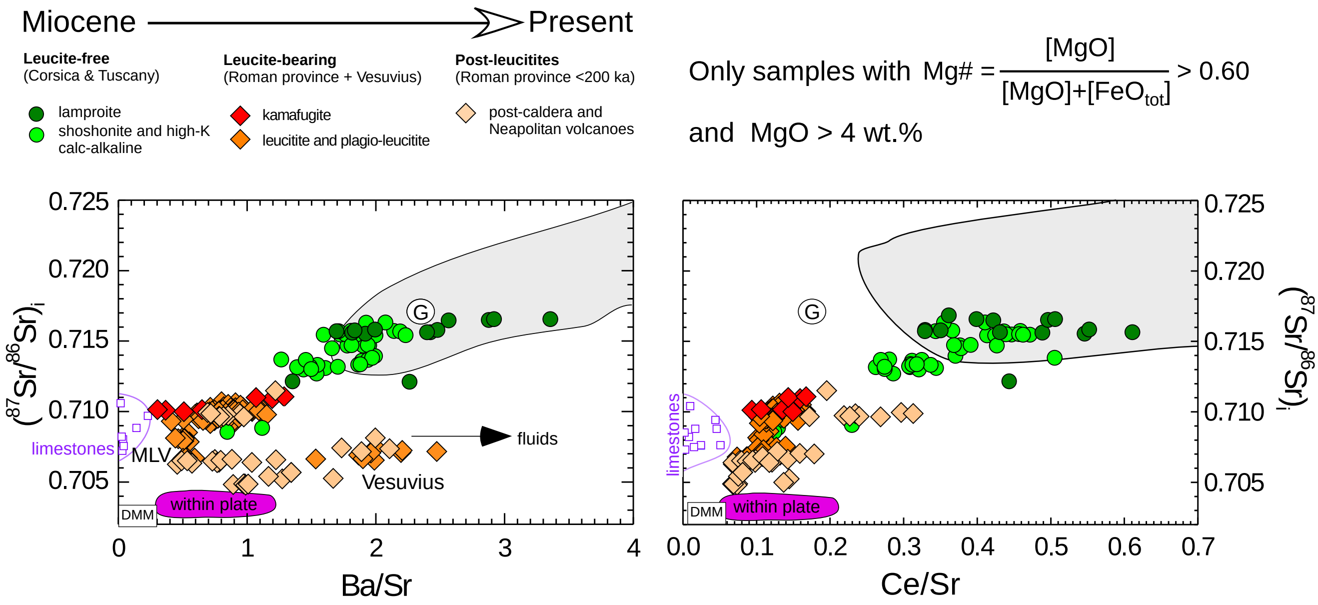

Figure 13 - Lithos - Conticelli et aal. (2014)
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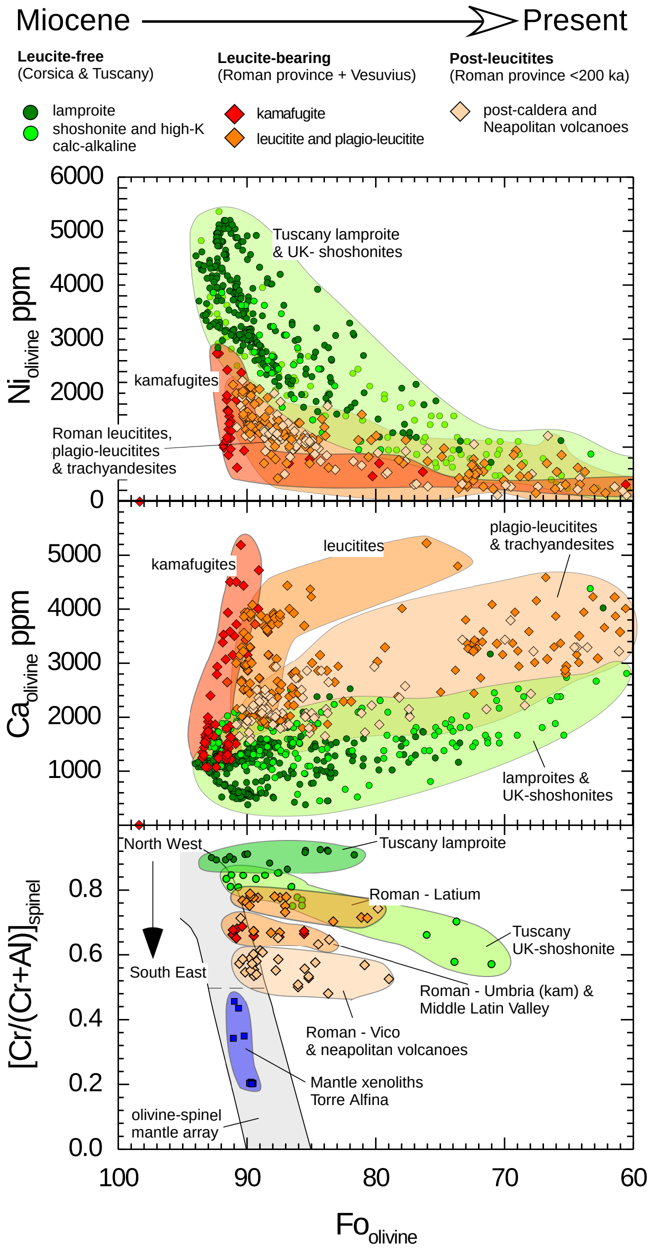

Figure 14 - Lithos - Conticelli et aal. (2014)
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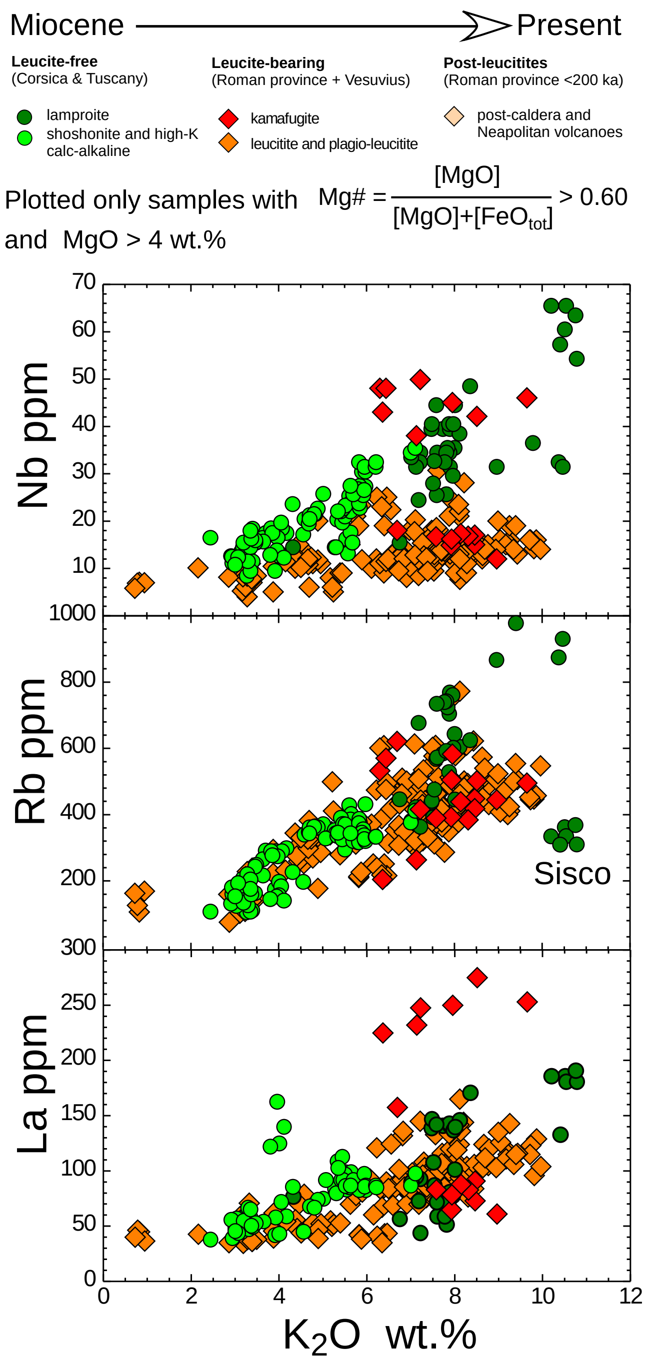

Figure 15 - Lithos - Conticelli et aal. (2014)
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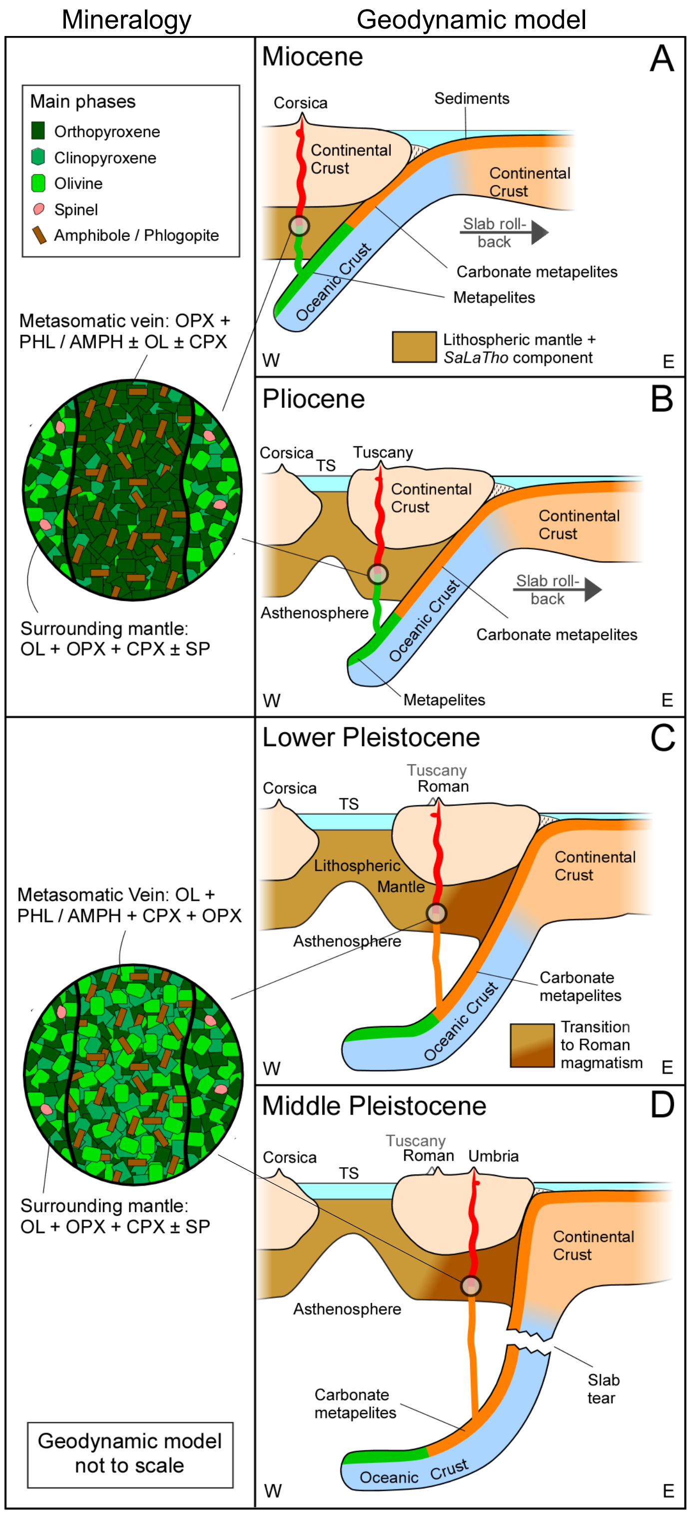

Mineralogy
Geodynamic model
Main phases
Orthopyroxene
Clinopyroxene
Olivine
Spinel
Amphibole / Phlogopite
Metasomatic vein: OPX + PHL / AMPH ± OL ± CPX
Surrounding mantle: OL + OPX + CPX ± SP
Metasomatic Vein: OL + PHL / AMPH + CPX + OPX
Surrounding mantle: OL + OPX + CPX ± SP
Geodynamic model not to scale
Miocene
A
Corsica
Sediments
Continental Crust
Continental Crust
Slab roll-back
Carbonate metapelites
Metapelites
Oceanic Crust
Lithospheric mantle + *SaLaTho* component
W
E
Pliocene
B
Corsica
TS
Tuscany
Continental Crust
Continental Crust
Slab roll-back
Asthenosphere
Oceanic Crust
Carbonate metapelites
Metapelites
W
E
Lower Pleistocene
C
Tuscany
Roman
Corsica
TS
Lithospheric Mantle
Continental Crust
Asthenosphere
Carbonate metapelites
Oceanic Crust
Transition to Roman magmatism
W
E
Middle Pleistocene
D
Tuscany
Roman
Umbria
Corsica
TS
Continental Crust
Asthenosphere
Slab tear
Carbonate metapelites
Oceanic Crust
W
E